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2	Urban forests near roads do not reduce gaseous air pollutant concentrations
3	but have an impact on particles levels
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24 Abstract

The ability of urban vegetation to improve air quality for the benefit of urban residents is often 25 considered fact since plants can absorb and capture air pollutants. However, there is little 26 empirical evidence that urban air quality at the local scale is improved by the presence of, e.g. 27 28 trees, especially in northern climatic regions. We studied the impact of urban forest vegetation 29 on the levels of five types of air pollutants (NO<sub>2</sub>, ground-level O<sub>3</sub>, anthropogenic and biogenic VOCs, and particulate matter) in near-road environments during summer (June) using passive 30 samplers in Helsinki, Finland. Concentrations of gaseous pollutants did not differ significantly 31 32 between tree-covered and adjacent open areas, while particle pollutant levels were significantly lower in tree-covered areas than in adjacent open, treeless areas. Vegetation-related variables 33 34 (canopy closure, tree number and size, and ground vegetation) did not explain differences in air 35 quality. Our results suggest that the role of urban, mostly deciduous, vegetation is negligible in improving local air quality, in terms of the anthropogenic pollutants measured here, in northern 36 climates. However, air particulate pollution, which is likely to be dominated by large-sized 37 particles in our study, can be reduced by urban vegetation. 38

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#### 40 Highlights:

Gaseous pollutant concentrations did not differ between tree-covered and open areas
Particle pollutant levels were significantly lower in tree-covered areas
Vegetation-related variables did not explain the difference in particulate levels

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# **1. Introduction**

49	Poor air quality due to air pollutants is amongst the most recognized environmental problems in
50	urbanized areas around the world. In Europe, the levels of air pollutants have generally
51	decreased within the past decades, but in many urban areas the levels of particulate matter,
52	nitrogen dioxide (NO <sub>2</sub> ), ground-level ozone (O <sub>3</sub> ), and anthropogenic volatile organic compounds
53	(AVOC) are still high enough to cause severe risk to human health (EEA, 2014). Most of these
54	air pollutants originate from energy production and road traffic (EEA, 2014).
55	
56	Although reducing air pollutant emissions is likely to be the most effective way to improve air
57	quality (EEA, 2014), it is widely believed that vegetation – due to its ability to absorb and
58	capture air pollutants with its large leaf area – can be used to mitigate urban air pollution
59	problems (Beckett, Freer-Smith & Taylor, 2000a; Nowak, 2006; Nowak, Crane & Stevens,
60	2006). For example, trees and herbaceous plants capture particulate pollution by dry deposition
61	on their large leaf surfaces (Hofman, Stokkaer, Snauwaert & Samson, 2013; Räsänen,
62	Holopainen, Joutsensaari, Ndam, Pasanen, Rinnan & Kivimäenpää, 2013; Weber, Kowarik &
63	Säumel, 2014).
64	

Furthermore, trees and other vegetation can absorb gaseous air pollutants, mainly from air to
leaf, where gases such as NO<sub>2</sub> (Chaparro-Suarez, Meixner & Kesselmeier, 2011; Rondón &
Granat, 1994; Takahashi, Higaki, Nohno, Kamada, Okamura, Matsui, Kitani & Morikawa,
2005), O<sub>3</sub> (Harris & Manning, 2010; Manes, Incerti, Salvatori, Vitale, Ricotta & Costanza, 2012;
Wang, Zhou, Wang, Gao, Zheng, Tong & Ouyang, 2012) and anthropogenic volatile organic

compounds (AVOC) (Doty, James, Moore, Vajzovic, Singleton, Ma, Khan, Xin, Kang, Park,
Meilan, Strauss, Wilkerson, Farin & Strand, 2007; Keymeulen, Schamp & Van Langenhove,
1995) are absorbed through the stomata into the leaf interior. Consequently, the mitigation of air
pollution is often considered an important ecosystem service provided by urban vegetation (e.g.
Chaparro & Terradas, 2009; Jim & Chen, 2008; Manes et al., 2012; Nowak, Crane, Stevens,
Hoehn, Walton & Bond, 2008).

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77 Forests and other vegetation in rural and urban areas emit biogenic volatile organic compounds 78 (BVOC) while human activities produce AVOCs, which, together with  $NO_x$  (NO + NO<sub>2</sub>), play a key role in the formation of ozone  $(O_3)$  in the tropospheric air (Calfapietra, Fares, Manes, 79 80 Morani, Sgrigna & Loreto, 2013; Loreto & Schnitzler, 2010). Under VOC-limited conditions, 81 ground-level  $O_3$  levels are usually lower in urban (high NO<sub>x</sub> levels) than in rural areas with NO<sub>x</sub>limited conditions (low NO<sub>x</sub> levels, but higher BVOC levels) (Calfapietra et al., 2013; EEA, 82 2014). However, in recent years, O<sub>3</sub> levels in European and North American cities have 83 increased more than in rural sites, although peak values are decreasing in both environments 84 85 (Paoletti, De Marco, Beddows, Harrison & Manning, 2014).

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Despite the wealth of studies suggesting that urban vegetation is able to purify air for the benefit
of urban inhabitants, contradictory results and comments have emerged recently (Gromke &
Ruck, 2009; Harris & Manning, 2010; Pataki, Carreiro, Cherrier, Grulke, Jennings, Pincetl,
Pouyat, Whitlow & Zipperer, 2011; Pataki, Alberti, Cadenasso, Felson, McDonnell, Pincetl,
Pouyat, Setälä & Whitlow, 2013; Vos, Maiheu, Vankerkom & Janssen, 2013). Critique is based
on the interpretation of model studies, according to which ambient air quality should uniformly

be improved by the presence of vegetation in the urban environment, where pollutant
concentrations are high (e.g. Baumgardner, Varela, Escobedo, Chacalo & Ochoa, 2012;
Hirabayashi, Kroll & Nowak, 2012; McDonald, Bealey, Fowler, Dragosits, Skiba, Smith,
Donovan, Brett, Hewitt & Nemitz, 2007; Morani, Nowak, Hirabayahsi & Calfapietra, 2011;
Nowak, Hirabayashi, Bodine & Hoehn, 2013).

Only a few studies exist in which pollutant levels have been measured locally, e.g. within a 99 forest or park canopy and compared to pollution levels in adjacent open areas (see Brantley, 100 101 Hagler, Deshmukh & Baldauf, 2014; Cavanagh, Zawar-Reza & Wilson, 2009; Freer-Smith, Beckett & Taylor, 2005; Harris & Manning, 2010; Setälä, Viippola, Rantalainen, Pennanen & 102 103 Yli-Pelkonen, 2013; Streiling & Matzarakis, 2003; Viippola, Rantalainen, Yli-Pelkonen, Tervo 104 & Setälä, 2016; Yin, Shen, Zhou, Zou, Che & Wang, 2011). As such, to enhance our understanding on the uptake, deposition, and re-suspension rates of pollutants by urban 105 vegetation, more on-site, small-scale measuring campaigns are needed (Pataki et al., 2011, 2013; 106 107 Whitlow, 2009).

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Our main objective is to explore the ability of urban vegetation to purify gaseous and particulate air pollutants of anthropogenic, mostly traffic-derived origin under northern summertime conditions in the near-road environment in Finland. This study builds on the same empirical principles as applied by Setälä et al. (2013), in which air pollutant levels (concentrations of NO<sub>2</sub> and VOCs and mass deposition of particles) were measured in open and tree-covered areas in late summer and winter using passive samplers in urban near-road environments in two cities (Helsinki and Lahti) in Finland. The present study was conducted during midsummer (mainly 116 June) when the total leaf-area and gas exchange between leaves and the ambient air is expected to be higher than in Setälä et al. (2013). The sampling protocol of the present study was also 117 slightly different by fine tuning the position of the sampling sites in relation to the prevailing 118 119 wind direction. Based on previous findings in similar environments (Setälä et al., 2013) we 120 hypothesized that concentrations of gaseous air pollutants should not differ between urban tree-121 covered and open, treeless areas in near-road environments, while air particulate levels (mass of all deposited particles) should be lower in such tree-covered areas. We also assumed that the 122 removal of pollutants, particularly air particulates, should relate to the volume and structure of 123 124 vegetation in the tree-covered areas. 125 2. Methods 126 127 128 2.1. Sampling methods 129 We measured air pollutant levels using dry deposition passive samplers, placed either under tree 130 canopies in tree-covered areas or in adjacent treeless open areas in near-road environments in the 131 Helsinki Metropolitan Area (60°10′15″N, 24°56′15″E), southern Finland (Fig. 1). The air 132 pollutants measured were i) nitrogen dioxide (NO<sub>2</sub>), ii) ground-level ozone (O<sub>3</sub>), iii) a selection 133 of typical anthropogenic volatile organic compounds (AVOCs), iv) biogenic volatile organic 134 135 compounds (BVOCs), which is a selection of the most common monoterpenes in Finnish forests alongside isoprenes (Lindfors & Laurila, 2000), and v) particulate matter. For NO<sub>2</sub> and O<sub>3</sub> we 136 137 used diffusive samplers developed by the Swedish Environmental Research Institute IVL, where the gas is adsorbed to a filter paper inside the collector and the amount of gas is analyzed by 138

extracting it from the filter to distilled water, after which the amount of gas is determined with a 139 spectrophotometer (Ferm & Svanberg, 1998). AVOCs and BVOCs (see Table 3 for the list of 140 compounds) were sampled using diffusive Carbopack B adsorbent tubes and analyzed according 141 to the EN ISO 16017-2 standard, where the concentrations of the studied compounds, counted 142 143 from individual adsorbent tubes, are determined by comparing them to compound-specific 144 standard substances. VOC results are presented as total AVOCs (= T-AVOCs, including all the sampled AVOC compounds) and as total BVOCs (= T-BVOCs, including all the sampled BVOC 145 compounds). Particle pollution levels were measured using passive collectors developed by IVL 146 147 (Ferm, Watt, O'Hanlon, De Santis & Varotsos, 2006). NO<sub>2</sub> samplers and their analyses were provided by Metropolilab, Helsinki, Finland,  $O_3$  and particle samplers and their analyses by IVL, 148 149 and VOC samplers and their analyses by Ramboll Analytics, Lahti, Finland.

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The sampling of  $NO_2$ ,  $O_3$  and VOCs is based on molecular diffusion. The method has some 151 limitations, but has been successfully used in numerous studies, with results strongly in line with 152 continuous air monitoring (Ayers, Keywood, Gillett, Manins, Malfroy & Bardsley, 1998; HSY, 153 2014; Krupa & Legge, 2000). The passive particle sampling method is based on the deposition of 154 155 particles by impaction and diffusion on a vertically-mounted cylindrical Teflon surrogate 156 surface. The method does not provide information on the specific particle size fractions, but provides the mass of deposited particles on the sampler surface. Mass is calculated by weighing 157 158 the surrogate surface before and after exposure. The surrogate surfaces are equilibrated for 24 h before weighing in a weighing room at 20°C and 50% relative humidity (Ferm et al., 2006). 159 160 Ferm et al. (2006) noted that particle deposition on Teflon correlated well with  $PM_{10}$ concentrations when samplers were not situated in the immediate vicinity of roads. 161

#### 163 2.2. Sampling sites and dates

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Ten sampling sites [different from those in Setälä et al. (2013)] in the Helsinki Metropolitan 165 166 Area (eight in the city of Helsinki, and one each in the neighboring cities of Espoo and Vantaa, 167 Fig. 1b) were established on the northern side of roads oriented in an east-west direction with moderate to heavy traffic flows (Table 1). This ensured that air pollutant collectors resided 168 downwind from traffic-derived air pollutants. There were no major intersections or roads 169 170 oriented in a south-north direction, or other roads oriented in a west-east direction to the north of the measuring sites that could potentially have biasing effects. Each of the 10 sites consisted of a 171 172 pair of sampling units placed on the northern side of the road: one unit in an open area and 173 another in a tree-covered area. The sampling sites were approximately level with the road surface. The open areas were meadows, grasslands or other treeless areas. The soil surface at 174 these open areas was either completely pervious or partly impervious with asphalt walking and 175 cycling paths. The tree-covered areas were mature urban forests of mainly broadleaved trees and 176 a rich understorey layer. Although all sites resided in the urban environment, no buildings or 177 178 other urban infrastructure existed in close proximity to the sites (see e.g., Fig. 1c).

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The samplers were mounted under rain shields fixed to 40 cm-long aluminium L-shaped sampler holders (together forming a sampling unit), which were attached to lamp posts or wooden poles in the open areas. In the tree-covered areas the sampler holders were attached to tree trunks directly under the canopy. The shields and sampler holders were manufactured by IVL. The sampling units for NO<sub>2</sub>, VOCs and particles were mounted 2.0-3.5 m above ground depending

185 on a suitable mounting structure.  $O_3$  sampling units were mounted 3.2 m above ground to prevent them from being too close to the ground where O<sub>3</sub> gets depleted (Mills, Pleijel, Büker, 186 Braun, Emberson, Harmens, Haves, Simpson, Grünhage, Karlsson, Danielsson, Bermejo & 187 Gonzalez Fernandez, 2010). Within each site, the sampling units (open and tree-covered) were 188 189 always placed at the same distance from the side of the road (a line marking the outer boundary 190 of the road). At different sites, depending on the availability of suitable mounting structures, the sampling unit pairs were situated at slightly different distances from the road side; ranging 191 between 18 and 30 m (mean = 25.6 m). The forest edge in tree-covered areas was, on average, 192 193 6.2 m (0 - 14 m) from the road side. The area between the road and the forest edge was open with short grass or meadow vegetation. Measurements were carried out during midsummer when 194 plant leaves were fully developed: NO<sub>2</sub>, VOCs and particle levels were measured from 29 May 195 196 to 26 June, 2013 (28 days) and  $O_3$  was measured from 2 June to 7 July, 2014 (35 days).

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A set of environmental variables was quantified at the sites (Table 1): Canopy closure at the tree-198 covered areas was estimated using two upwards facing photographs taken 1 m above ground. 199 200 One photograph was taken underneath the sampling unit and the second one 5 m towards the 201 road from the sampling unit. The two photographs represented canopy closure well enough due to the rather homogenous tree/canopy composition within each forested site. The proportion of 202 non-visible sky, a proxy for canopy closure, was calculated using Adobe Photoshop image 203 204 processing software. The number and size of trees [including trees with a diameter at breast height, DBH > 2.54 cm (= 1 inch)], and whether the trees were deciduous or coniferous, were 205 206 determined from a sector covering 90° from the sampling unit perpendicular towards the road 207 that delineated the area. The number of trees was converted to represent number per unit area

(100 m<sup>2</sup> in this case). The extent of ground vegetation (shrubs, bushes, meadow vegetation) was
estimated visually at both tree-covered and open areas and classified into dense, intermediate and
meager (Table 1).

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The degree of canopy closure was  $84.7\% \pm 8.0$  (mean  $\pm$  SD) in the tree-covered sampling sites. 212 The total number of trees recorded was  $73.7 \pm 45.5$ , with large trees (DBH > 32 cm) comprising 213 4.4% ( $\pm$  5.1) of all the trees. The sampling sites were dominated by deciduous trees (96%  $\pm$  7). 214 including forest tree species (*Betula* spp. and *Sorbus aucuparia* being dominant with scattered 215 *Populus* spp. and *Prunus padus*) typical to Southern Finland. Traffic flow data (monthly average, 216 Table 1) were obtained from the City of Helsinki (2012) and the Finnish Transport Agency 217 (2010). Mean annual anthropogenic pollution concentrations, measured at several sampling 218 locations in Helsinki in 2013, were 14–49  $\mu$ g m<sup>-3</sup> for NO<sub>2</sub>, 39–52  $\mu$ g m<sup>-3</sup> for O<sub>3</sub>, 2–6  $\mu$ g m<sup>-3</sup> for 219 AVOCs (benzene, toluene and xylenes combined) and  $11-25 \ \mu g \ m^{-3}$  for PM<sub>10</sub> (Air quality in 220 Finland, 2015). Mean annual NO<sub>2</sub>, O<sub>3</sub> and PM<sub>10</sub> concentrations at municipal monitoring stations 221 do not usually exceed annual human health limit values of 40  $\mu$ g m<sup>-3</sup>, 120  $\mu$ g m<sup>-3</sup> and 40  $\mu$ g m<sup>-3</sup>, 222 respectively, with the exception of NO<sub>2</sub> in a few highly trafficked locations. Regarding AVOCs, 223 an annual threshold value is available only for benzene (5  $\mu$ g m<sup>-3</sup>). For BVOCs no threshold 224 values are available. A wind rose showing the prevailing wind direction and speed during the 225 measuring periods is shown in Fig. 2. The monthly average temperature in Helsinki in June 2013 226 227 was 18.1 °C and in June 2014 13.6 °C, representing typical temperatures in June in the Helsinki 228 area.

229

230 2.3. Data analysis

We analyzed the air pollution data using paired samples *t*-test to determine if there is a difference 232 in pollution levels between the tree-covered and open study areas. We considered *p*-values < 233 0.05 to indicate statistically significant differences between treatments. Data were  $\log_{10}$ 234 transformed to stabilize the variance of individual properties where necessary. Linear regression 235 was applied to detect possible correlations between measured environmental variables and 236 pollution level differences between the tree-covered and open areas. In open areas, the influence 237 of ground vegetation on air pollution was tested using the pollution levels of open areas. T-238 239 BVOCs were not tested because BVOCs are emitted mainly by trees and not by shrubs and other typical grassland vegetation. The impact of traffic flow on pollution levels was tested using the 240 actual pollutant levels measured at each sampling point, including both open and tree-covered 241 242 areas. T-BVOCs were not tested because BVOCs are not traffic-emitted substances. Statistical analyses were performed using SPSS Statistics, version 21. 243

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#### 245 **3. Results**

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Results for each air pollutant are presented both as means ( $\pm$  SD) of all sites (tree-covered vs. open area comparisons), and for each site separately along the traffic flow gradient (number of motor vehicles day<sup>-1</sup>) to explore a potential correlation between pollutant levels and traffic flow (Fig. 3). As for total BVOCs (T-BVOCs), values are presented in relation to % canopy closure to depict the association between increasing canopy closure and T-BVOC levels (Fig. 3). Concentrations of the gaseous pollutants did not differ significantly between tree-covered and open areas (NO<sub>2</sub>, 2.6% higher in tree-covered areas, *p* = 0.407; O<sub>3</sub>, 3.8% lower in tree-covered 254 areas, p = 0.177; total AVOC (T-AVOC), 2.7% lower in tree-covered areas, p = 0.718; T-BVOC, 9.5% higher in tree-covered areas, p = 0.432) (Fig. 3a-d). Thus, tree-cover had no effect on the 255 concentrations of gaseous pollutants, while traffic flow explained most of the variation in NO<sub>2</sub> 256 257 concentrations and some of the variation in O<sub>3</sub> concentrations between sites (Table 2). For T-AVOCs and particle levels, no such correlation was evident, neither in tree-covered nor in open 258 areas (Table 2). A detailed listing of the constituent AVOCs and BVOCs and their proportions 259 are shown in Table 3. The proportions of AVOC and BVOC constituents were insensitive to the 260 presence of trees (results not shown). 261 262

In contrast to gaseous pollutants, particle pollution levels differed statistically significantly between the tree-covered and open study areas: particle levels were, on average, 23% (p = 0.023) lower in tree-covered areas than in open areas (Fig. 3e). None of the measured vegetation properties correlated statistically significantly with the difference in particle levels between the tree-covered and open areas (Table 2). As NO<sub>2</sub>, O<sub>3</sub>, T-AVOC and T-BVOC concentrations did not differ statistically significantly between tree-covered and open areas, the relationships between their differences and vegetation properties could not be tested.

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None of the vegetation properties measured in the tree-covered areas correlated statistically significantly with T-BVOC levels (results not shown), although there appears to be a trend in T-BVOC levels for total number of trees (R = 0.616, p = 0.078) and % canopy closure (R = 0.546, p = 0.129).

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276 **4. Discussion** 

Results from our study, conducted under northern summertime conditions (June), suggest that 278 forest vegetation in near-road urban environments does not improve local air quality in terms of 279 gaseous (NO<sub>2</sub>, O<sub>3</sub>, AVOCs) air pollutants, thus corroborating earlier findings in late summer 280 281 (August-September) in two Finnish cities (Setälä et al., 2013). In a similar study in Sweden, but 282 only from one measurement site, Grundström and Pleijel (2014) found slightly (7%) reduced NO<sub>2</sub> concentrations in the forest canopy. However, besides the canopy effect, this reduction may 283 have resulted from the 4 m difference in distance between their open and canopy sampling points 284 285 from a heavy traffic road (the latter being further away from the road) (see e.g. HSY, 2014). In the same study, the authors did not detect reduced  $O_3$  concentrations in the canopy (Grundström 286 & Pleijel, 2014), which is in line with our results. Fantozzi, Monaci, Blanusa & Bargagli (2015) 287 288 found lower NO<sub>2</sub> concentrations under the canopies of Mediterranean trees (evergreen Quercus ilex L.) growing 1-10 m from a road compared to a nearby open-field transect. However, reduced 289 O<sub>3</sub> concentrations in the canopy were observed only during post-summer rainfalls (Fantozzi et al. 290 2015), indicating that the impact of vegetation on gaseous pollution concentrations may also 291 292 depend on the type of local vegetation and climatic conditions. On the other hand, Harris and 293 Manning (2010) found, also using passive samplers, higher NO<sub>2</sub> and lower O<sub>3</sub> levels within urban tree canopies than outside them. They suggested that this results from  $NO_x/O_3$  chemistry 294 related to gas interactions between soil and the air, as described by Fowler (2002). Although the 295 296 three gases in our study were sampled at the same locations, O<sub>3</sub> was collected a year later than NO<sub>2</sub> and VOC preventing us from comparing NO<sub>2</sub> and VOC levels directly with those of O<sub>3</sub>. 297 298 Furthermore, using data collected during one month only to unravel complex interactions

between NO<sub>2</sub>, O<sub>3</sub> and VOCs is challenging due to the rapid and complex dynamics of these
compounds in the air.

302	Our results, however, do support earlier studies showing that trees can mitigate problems
303	associated with particulate air pollution (e.g. Beckett et al., 2000a,b; Cavanagh et al., 2009;
304	Tallis, Taylor, Sinnett & Freer-Smith, 2011), particularly in the near-road environment (Baldauf,
305	Thoma, Khlystov, Isakov, Bowker, Long & Snow, 2008; Islam, Rahman, Bahar, Habib, Ando &
306	Hattori, 2012; Maher, Ahmed, Davison, Karloukovski & Clarke, 2013; Setälä et al., 2013;
307	Steffens, Wang & Zhang, 2012), although the tree effect can depend on environmental
308	conditions such as wind direction in relation to pollution source, as well as particle size (Brantley
309	et al., 2014; Hagler, Lin, Khlystov, Baldauf, Isakov, Faircloth & Jackson, 2012; Tong, Whitlow,
310	MacRae, Landers & Harada, 2015). Although the method used in our study does not allow for
311	the measurement of size fractions of the collected particles, it is likely that coarse particles, most
312	likely road dust, were emphasized in the mass deposition (Ferm et al., 2006); the Teflon
313	sampling surfaces were visibly dirtier close to the road than further away. However, regarding
314	smaller particles, the situation can be different: Tong et al. (2015) investigated the dispersion of
315	particulates near roads using aerosol spectrometers in brief monitoring campaigns and found that
316	$PM_{2.5}$ concentrations were higher along a transect with trees downwind from the road and
317	declined less sharply than along open transects without trees. Also, as noted by Setälä et al.
318	(2013), particle levels, measured with passive samplers, were notably and significantly lower in
319	tree-covered areas compared to open areas, but often inconsistent with particle data gathered
320	using "active" collecting devices, which can specify particle size fractions and produce results
321	both in mass and number. Considering the particle deposition results and the measuring

methodology in this study, it should be noted that particles that can enter throughout the human
respiratory system are smaller than 2.5 µm in diameter, and these are easily outweighed by larger
particles.

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326 Sampling in the current study was done in June, i.e. during midsummer in Finland when total 327 leaf area, as well as gas exchange between leaves and ambient air, is high (Rautiainen, Heiskanen & Korhonen, 2012). However, when comparing our results to Setälä et al. (2013), in 328 which sampling took place in late summer (from early August to early September) when leaf 329 330 activity is ceasing, it is interesting to note that road-side vegetation did not reduce  $NO_2$  and AVOC concentrations neither during mid- nor late summer seasons. In order to explore the 331 canopy effect for the entire growth period, the duration of measuring campaigns during the same 332 year should extend over the entire leaf period. 333

334

Estimates on the ability of trees to absorb anthropogenic VOCs are based on a few laboratory 335 incubation studies (Cornejo, Munoz, Ma & Stewart, 1999; Keymeulen et al., 1995; Yang, 336 Pennisi, Son & Kays, 2009). To our knowledge, the fate of AVOCs in relation to urban 337 338 vegetation has only been investigated in one study in Las Vegas, U.S. (Hiatt, 1998), where samples of air and leaves of several tree species were analyzed. Hiatt (1998) showed that several 339 local tree species can absorb AVOCs according to existing models. However, our results indicate 340 341 that the potential of trees to create areas of lower AVOC concentrations at the local scale in northern climates is meager, at least in urbanized sites where deciduous trees constitute most of 342 343 the vegetation, corroborating Setälä et al. (2013).

Concentrations of BVOCs appeared to be relatively low at most of the study sites (often barely above the detection limit), and were not significantly different between the open and treecovered areas. Although conifers in Nordic regions are efficient emitters of monoterpenes, some deciduous tree species, such as *Betula pendula*, *B. pubescens*, *Populus tremula* and *Salix* sp., are also known to emit monoterpenes (Hakola, Rinne & Laurila, 1999; Lindfors & Laurila, 2000). Deciduous trees made up most of the canopy cover at our study sites, but BVOC concentrations were equally low in most of the open and tree-covered sites.

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353 That concentrations of gaseous pollutants appeared to be unresponsive to forest vegetation, while particle pollution levels were clearly reduced by tree-cover, can be due to the inefficiency of 354 northern vegetation to absorb and process gaseous pollutants, such as NO<sub>2</sub>, O<sub>3</sub> and AVOCs. The 355 356 amount of pollutants absorbed by stomatal intake seems to be irrelevant in the context of ambient concentrations. However, even if the vegetation could absorb these pollutants efficiently, it is 357 possible that reduced air flow within tree-covered sites (Belcher, Harman & Finnigan, 2012; 358 Gromke & Ruck, 2009; Wuyts, Verheyen, De Schrijver, Cornelis & Gabriels, 2008) can increase 359 360 pollutant levels within the canopy and thus have negative impacts on local air quality (see Setälä 361 et al., 2013; Viippola et al., 2016; Vos et al., 2013), while in open areas the polluted air mass is mixed by wind and diluted more rapidly. Such an indirect effect of vegetation on local air quality 362 363 is seldom reported in the literature and merits more research.

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The clear reduction in particle levels at tree-covered sites did not relate to any of the measured vegetation properties. It is possible that the vegetation parameters measured here were not instrumental in creating potential variation in air particulate levels and more parameters would have been needed to tackle this variation. Nevertheless, the reduction in particle levels is best
explained by either effective deposition of large (and heavy) particles on vegetation surfaces or
perhaps - more importantly - by reduced ventilation under the canopy. Wind speed is known to
be lower under thick canopies compared to open, treeless areas (Belcher et al., 2012; Setälä et
al., 2013; Wania, Bruse, Blond & Weber, 2012), which likely prevents road dust from dispersing
further away from the road in tree-covered areas.

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Due to the fact that air pollution patterns can be shaped by wind conditions in urban areas (Choi, 375 376 He, Barbesant, Kozawa, Mara, Winer & Paulson, 2012; Kozawa, Winer & Fruin, 2012; McNabola, Broderick & Gill, 2009), the placement of sampling devices in relation to prevailing 377 wind patterns deserved attention in our study set-up. As the prevailing wind direction (long-term 378 379 average) in June in Helsinki is from the south or south-west, all sampling sites were placed downwind from the pollution source. Wind direction, however, varied considerably during the 380 one-month measuring periods in 2013 and 2014 and no clear consistency in wind direction at the 381 study region was evident (Fig. 2). However, south-western and eastern winds dominated, and 382 only a small percentage of winds blew from the north in June 2013. Thus, for most of that 383 384 campaign period, our sampling units were downwind or at least in a neutral position, but clearly not upwind, in relation to pollution sources. In June 2014, when O<sub>3</sub> was measured, winds blew 385 from almost all directions. Moreover, the relatively long sampling period used is bound to 386 387 diminish the impacts of short-term wind direction changes and local eddies on our results. The concentrations of NO<sub>2</sub>, O<sub>3</sub> and AVOCs were in line with the available mean annual and monthly 388 389 (June 2013) concentration data from Helsinki, and always below the limit values for human 390 health (Air quality in Finland, 2015; HSY, 2014).

### **5.** Conclusions

The current study conducted in Helsinki corroborates results from other studies in Nordic urban environments (Setälä et al., 2013; Viippola et al., 2016); the impact of the tree canopy, at least in near-road environments without buildings, is negligible regarding the concentrations of certain gaseous air pollutants. Alternatively, it may be that these gaseous pollutants were absorbed to some extent by the canopy, but not sufficiently so to compensate for increased concentrations due to reduced air-flow in tree-covered areas (see Janhäll, 2015). However, the levels of coarse particulate matter were reduced by the presence of trees. The methodology used does not allow for the extrapolation of this conclusion to fine particles. Our results - obtained from the period when gas exchange between foliage and the atmosphere is most active - thus suggest that forest vegetation does not necessarily reduce air pollutant levels in urban areas. However, further empirical field research on the mechanisms and factors contributing to the various direct and indirect impacts of vegetation on air pollutant levels is needed. Acknowledgments 

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## LIST OF TABLES

**Table 1.** Environmental variables measured at the 10 study sites in the Helsinki Metropolitan Area arranged by ascending traffic flow (number of motor vehicles day<sup>-1</sup>, monthly average for June). Values referring to canopy closure, trees and ground vegetation were estimated from a sector covering 90 degrees from the sampling unit towards the road that delineated the area. Trees with a diameter at breast height (DBH) of < 2.54 cm (= 1 inch) are not included.

	Canopy	Total nr. of	Nr. of trees with DBH	Tree-covered area	Open area -	Traffic
Site	closure (%)	trees 100 m <sup>-2</sup>	$> 32 \text{ cm } 100 \text{ m}^{-2}$	- ground vegetation	ground vegetation	flow
1	71	13.0	0	dense	dense	8 000
2	75	7.0	0.3	intermediate	dense	14 100
3	95	19.4	1.2	meager	intermediate	14 700
4	82	17.6	0.6	intermediate	intermediate	14 800
5	89	7.4	0.4	intermediate	dense	52 600
6	94	3.3	0.6	intermediate	dense	58 500
7	88	13.8	0	meager	intermediate	64 100
8	78	7.8	0	intermediate	meager	65 000
9	89	5.8	0.5	dense	dense	91 600
10	87	21.4	0.2	intermediate	intermediate	102 400

**Table 2.** Relationship between vegetation properties and the statistically significant air quality difference of particle levels\*. The difference is calculated by subtracting particle levels detected in the tree-covered areas from that in the open areas. Linear regression was used to detect relationships and their statistical significances. The influence of ground vegetation of the open areas was tested using the pollution levels of the open areas (except for T-BVOCs that are mainly emitted by trees). The impact of traffic flow on pollution levels (except for T-BVOCs that are not traffic-emitted) was tested using the actual pollutant levels measured at each sampling point, including both open and tree-covered areas. Statistically significant *R* values are marked in bold.

\* = Regarding NO<sub>2</sub>, O<sub>3</sub>, T-AVOCs and T-BVOCs, the differences between tree-covered and open areas were not statistically significant, and thus the relationships between their differences and vegetation properties were not tested (reason for blank lines). <sup>a</sup> n = 10.

<sup>b</sup> For traffic flow, n = 20, except for T-AVOC (n = 19).

		Canopy	Total nr of trees $100 \text{ m}^{-2}$ a	Nr of trees with DBH $\sim 22 \text{ am } 100 \text{ m}^{-2} \text{ a}$	Tree-covered area's ground	Open area's ground	Traffic flow <sup>b</sup>
		closule		> 32 CIII 100 III	vegetation	vegetation	
P-dep.	R	0.023	0.484	0.090	0.036	0.138	0.024
	р	0.950	0.157	0.804	0.921	0.704	0.920
$NO_2$	R	_	_	_	_	0.130	0.787
	р	_	—	_	_	0.720	<0.001
O <sub>3</sub>	R	_	_	_	_	0.074	0.545
- 5	p	_	_	-	_	0.839	0.013
T-AVOC	R	_	_	_	_	0 169	0.207
	p	_	_	_	_	0.642	0.395

**Table 3.** List of anthropogenic (AVOC) and biogenic (BVOC) volatile organic compounds and their relative proportions in the samples. Values shown are mean proportions of all the samples collected at the tree-covered and open areas.

AVOC	proportion
Toluene	34 %
Benzene	12 %
Ethylbenzene	7.7 %
o-xylene	9.7 %
<i>m/p</i> -xylene	22 %
Styrene	2.3 %
Ethyl tert-butyl ether	0.9 %
Propylbenzene	1.6 %
2-Ethyltoluene	2.9 %
3-Ethyltoluene	5.2 %
4-Ethyltoluene	1.8 %
BVOC	
a-pinene	47 %
b-pinene	13 %
d-carene	13 %
limonene	26 %



**Fig. 1.** (a) A map of Finland with (b) an aerial image of the Helsinki Metropolitan Area displaying locations of the ten sampling sites (1 - 10), and (c) an aerial image of one of the sampling sites (site 5) as an example. At each of the 10 sites, air quality was measured in one tree-covered and one open area. At site 5, sampling units were placed ca. 30 m from the road edge.



**Fig. 2.** Wind direction and speed during 29 May – 26 June, 2013 and 2 June – 7 July, 2014 in Helsinki (Kumpula measuring station). Data were provided by the Finnish Meteorological Institute (FMI).



0,000,000,000,000,000

- 12<sup>60</sup> 13<sup>60</sup>

Traffic flow

800, 410, 410, 480 5

**Fig. 3.** Concentrations of (a) NO<sub>2</sub>, (b) O<sub>3</sub>, (c) total AVOCs, (d) total BVOCs and (e) levels of particles (mass of airborne particles deposited on the sampler) in open (white bars) and tree-covered (grey bars) areas. The first pair of bars in each panel refers to mean  $\pm$  SD (n = 10, except for total AVOCs and total BVOCs where n = 9 due to a lost VOC sample in one tree-covered study area) pollution levels of all 10 study sites. The rest of the bar pairs describe each study site separately. The sites are presented in relation to increasing traffic flow (number of motor vehicles day<sup>-1</sup>), except for T-BVOCs (panel d), where data are presented in relation to % canopy closure.