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► **To cite this version:**

Katell Quenea, Iry Andrianjara, Aleksandar Rankovic, Erika Gan, Emmanuel Aubry, et al.. Influence of the residence time of street trees and their soils on trace element contamination in Paris (France). Environmental Science and Pollution Research, 2019, 26 (10), pp.9785-9795. 10.1007/s11356-019-04405-w . hal-02164653

HAL Id: hal-02164653

<https://hal.sorbonne-universite.fr/hal-02164653>

Submitted on 25 Jun 2019

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Influence of the residence time of street trees and their soils on trace

element contamination in Paris (France)

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Abstract

With the actual increasing interest for urban soils, the evaluation of soil contamination by trace elements and the dynamics of this contamination appear mandatory to preserve plant and thereby human health. Street trees and the associated soil placed in pits located nearby roads could represent convenient indicators of urban and vehicle traffic influences on soils and plants. However, data on these soils remain scarce, many studies investigating park soils rather than street tree soils. Furthermore, trace elements could be one of the main factors causing the

29 observed urban tree decline, while practitioners more and more question the possible reuse of these soils after the
30 death of trees as well as tree litter collected in the streets. We evaluated the contamination in anthropogenic trace
31 elements (TE), namely Zn, Pb and Cd, of street trees (*Tilia tomentosa*) and their soils distributed all over Paris
32 (France). Street tree soils are imported from rural areas at the plantation of each new tree so that tree age
33 corresponds to the time of residence of the soil within an urban environment allowing the evaluation of temporal
34 trends on TE concentration in soils and trees.

35 The TE concentration revealed an important soil pollution, especially for the older soils (mean age of 80 years
36 old). The consideration of the residence time of trees and soils in an urban environment evidenced an
37 accumulation of Zn and Pb (*ca.* 4.5 mg kg⁻¹ yr⁻¹ and 4 mg kg⁻¹ yr⁻¹ for Zn and Pb, respectively). However, leaf
38 concentrations in TE were low and indicate that soil-root transfer was not significant compared to the
39 contamination by atmospheric deposition. These results underlined the necessity to deepen the evaluation of the
40 recycling of urban soils or plants submitted to urban contamination.

41

42 **Keywords:** Urban soils • Road traffic impact • Bioaccumulation • Trace element • Leaves • Roots

43

44 **Introduction**

45 For decades, trace elements (TE) have been of main concern in environmental survey due to their impact on
46 biodiversity and human health. In urban areas, heavy road traffic, industrial activities and residential heating
47 generate important atmospheric pollution leading to TE deposition (Galloway et al. 1982; Garnaud et al. 1999;
48 Manta et al. 2002; Basioli et al. 2006; Wong et al. 2006; Thevenot et al. 2007; Schreck et al. 2012). In addition,
49 direct inhalation of contaminated atmosphere or street dust enriched in TE by urban inhabitants has deleterious
50 effects on health (Pena-Fernandez et al. 2014). The atmospheric deposition is also source of soil contamination
51 and thereafter of river and groundwater contaminations, through the leaching of impervious urban surfaces.
52 Finally, TE contamination from the atmosphere to soils and waters induces contamination of plants through
53 atmosphere-plant transfer as well as soil- plant transfer (Chojnacka et al. 2005; Kabata Pendias, 2010).

54 Many studies have focused on TE in urban environments either directly by soil concentration measurements (Ge
55 et al. 2000; Manta et al. 2002; Maher et al. 2008) or through biomonitoring (Harmens et al. 2010; Deljanin et al.
56 2016; Gillooly et al. 2016). The use of vegetation as bioindicator of pollution is widely applied, plants being
57 relevant integrator of their environment (Markert et al. 2003). Thus, measurements of TE concentrations in

58 plants allow the assessment of the overall TE availability to plants without any distinction of the source
59 (atmosphere, soil, water). However, all other things remaining equal (in particular, soil properties) TE
60 concentrations in trees are influenced by tree species (Piczak et al. 2003; Pulford and Watson 2003), the precise
61 localization of the street trees in the city (Maher et al. 2008) and the TE considered and its speciation (Kabata-
62 Pendias 2004). Metals such as lead (Pb) and cadmium (Cd) accumulate close to intense traffic roads and their
63 concentrations decrease exponentially with increasing distance to the road (Viard et al. 2004, Nabulo 2006,
64 Werkenthin et al. 2014). Mosses and trees are frequently used in biomonitoring context (Markert et al. 1996;
65 Piczak et al. 2003; Baycu et al. 2006; Gratani et al., 2008, Anicic et al. 2011; Sawidis et al. 2011; Guéguen et al.
66 2012, Natali et al. 2016). However, soil-plant transfer has often been neglected. In consequence, the
67 simultaneous analysis of soil and plant present on a same site could help circumscribing limitations occurring
68 when plants or soils are used separately for the evolution of site contamination (Mertens et al. 2005).

69 The city of Paris (France) is the fifth most important city in Europe in term of population, with a high
70 density (20000 inhabitants per km²). Human activities contribute to a large part of TE in Paris through industry
71 emissions, vehicle exhaust, and residential heating. Despite a large decrease of TE deposition since 1994 (Azimi
72 et al. 2005a), this atmospheric deposition remains the main source of TE, as evidenced by Garnaud et al. (1999)
73 and Azimi et al. (2005a, b). However another source of TE in urban soil is the runoff water from streets and
74 roofs. In Paris, roof runoff was evidenced to be a significant source of Cd, Zn, and Pb (Garnaud et al. 1999;
75 Gromaire et al. 2002; Rocher et al. 2004) and led to an increase of TE in the Seine river basin (Thevenot et al.
76 2007). Nonetheless, extensive data on soil contamination by TE close to roads in Paris are still missing. The
77 study of street trees growing in the vicinity of car exhaust and their soil could allow better assessing the extent of
78 TE contamination in Paris. Indeed, atmospheric deposition, or street and roof leaching likely impact soils from
79 those street trees and trees could be contaminated directly by either atmospheric deposition or by soil-plant
80 transfer.

81 In Paris as in many cities, street trees are planted in pits that have been filed with agricultural soil
82 transported from the country side (Paris Green Space and Environmental Division, pers. comm.). The soil is
83 removed and replaced each time a tree dies and a young one is planted. This procedure provides a unique
84 opportunity to assess the contamination dynamics through the comparison of trees with various ages (until 80
85 years) and their corresponding soil. Indeed, the age of street trees is also the residence time of the soil within the
86 urban environment (Kargar et al. 2013). In addition, while TE accumulate during the whole tree life in their
87 soils, TE accumulate in roots and leaves only during the life span of these organs. Thus, investigating root and

88 leaf TE concentrations might allow distinguishing between the atmospheric or soil sources depending on the
89 intensity of TE translocation between roots and leaves. Indeed, if translocation is low, root TE will mainly come
90 from soil-plant contamination and leaf TE from atmosphere-plant contamination. In addition, leaves record the
91 contamination during a single leafy season whereas roots accumulate TE for several years depending on their
92 size and life span (Withington et al. 2006). In addition, practitioners are so far dumping the soil of dead trees
93 with the belief that these soils are no longer fertile, partially due to contamination by different sources of
94 pollution. The same thing goes for street tree litter collected as it is considered as waste in the EU legislation
95 (Nurmatov et al. 2016). Consequently, the evaluation of the soil and leaf contamination could help deciding
96 whether street tree soils and leaves should be reused or dumped after tree death and litter collection.

97 Therefore, our objectives were to evaluate: (i) the TE (Zn, Cd, and Pb) contamination of soils highly
98 exposed to traffic contamination and imported to Paris between 15, 50 and 80 years ago, and (ii) the
99 contamination of roots and leaves from the corresponding street tree (*Tilia tomentosa* Moench) and thus (iii) the
100 potential soil-plant transfer of these TE. The three model TE were chosen because of their frequently observed
101 anthropogenic origin reported in urban soils as well as their ecological importance: Zn is essential for plant
102 nutrition while Cd and Pb are not, and while Cd and Zn are mobile in soils and available for plants, Pb tend to
103 accumulate mainly in roots and remain at leaf surface (Madejón et al. 2004). The model tree was chosen because
104 it is one of the first dominant species in the streets of Paris.

105 **Materials and methods**

106 **Study area and sampling procedure**

107 Thirty roadside linden trees (*Tilia tomentosa* Moench) were selected as these trees are widespread in Paris
108 (France) (48.8534°N; 2.3488°E). The selected roadside trees were gathered in three age classes: ten trees
109 belonged to the “young” class (between 11 and 17 years old, mean age of 13 years old), ten trees to the
110 “medium” class (between 41 and 67 years old, mean age of 49 years old), and ten trees to the “old” class (69 and
111 86 years old, mean age of 80 years old). Tree age was determined by dendrochronological methods by David et
112 al. (2018). When a new tree is planted, a pit of about 3-4 m³ is dug and filled with imported soil from
113 surrounding peri-urban agricultural areas. Only vigorous trees with either bare or drain-covered soils were
114 selected to avoid important differences in terms of rooting conditions and water availability (Rahman et al.
115 2011).

116 Sampling occurred in July 2011 (Fig. 1). Soils were sampled in pits from roadside and roots were isolated from
117 the soil cores. Leaf samples were collected on trees grown in the corresponding pits. Trees were distributed all
118 over Paris, with trees in nearly all Paris districts, to ensure the representativeness of the sampling. Soil samples
119 were collected with an auger in the 10–30 cm horizon depth in pits of the tree selected. For each street tree pit,
120 two cores were sampled and then the composite sample was freeze-dried. Thereafter, samples were sieved
121 (<2 mm) discarding coarse plant residues and roots (around 1mm diameter) were collected. Shadow leaves were
122 cut at minimum 2 m height from all sides of the crown of the trees from which soil samples have been collected.

123 **Sample treatment procedures**

124 Leaves and roots were washed twice with deionized water in ultrasonic bath to remove any particle presents on
125 the surface, and finally dried and grinded.

126 Pseudo-total metal concentrations were measured after soil mineralization. In a first step, soil samples were
127 mineralized in aqua regia (mixture of 1/3 HNO₃ 70% and 2/3 HCl 37%) using a temperature-controlled digestion
128 system (DigiPREP Jr instrument, SCP Science, Baie-d'Urfé, Canada) at 120°C for 8 h and dried.

129 Leaves and roots were mineralized according to the following procedure: leave and root samples were placed in
130 Teflon flask with HNO₃ 70% for 24 h at 120 °C with a DigiPREP instrument. After cooling H₂O₂ 30% was
131 added and Teflon flasks placed at 120 °C for 24 h.

132 **Physico-chemical analyses**

133 Main physico-chemical characteristics of the soil samples are reported in Table 1 and indicate a relative
134 homogeneity in the soil characteristics.

135 Total organic carbon and nitrogen contents were measured using an elemental analyzer (Carbo Erba instrument
136 CHN NA 1500 series 2, Milan, Italy).

137 CaCO₃ content, pH (H₂O), cation exchange capacity (CEC; Metson method), and soil texture results were
138 provided by a soil analysis laboratory (INRA, Arras, France) according to standardized French procedures
139 (AFNOR NF ISO 10693, NF ISO 10390, NF X 31-130, NF X 31-107, respectively).

140 Zinc concentrations were measured by inductively coupled plasma-optical emission spectrometry ICP-OES
141 (instrument JY2000), whereas Pb and Cd concentrations were measured by inductively coupled plasma-mass
142 spectrometry ICP-MS (X Series II, Thermo Electron). Ten blank samples were added to the sequence following

143 the same treatment for method control. Each sample was analyzed in triplicate. The detection limits of Pb, Cd,
144 and Zn were 0.3, 0.2, and 2.2 $\mu\text{g L}^{-1}$, respectively, whereas the limits of quantification were about 0.4, 0.3, and
145 3 $\mu\text{g L}^{-1}$, respectively. The accuracy of calibrations was checked using a certified reference material (TMDA-
146 64.2) from Environment Canada. The concentrations found corresponded to the certified values $\pm 5\%$.

147 **Data treatment and statistical analysis**

148 Data processing and statistical analyses were performed with RStudio 1.0.153 (RStudio Inc., Boston,
149 Massachusetts, USA) using R 3.4.1 (R Foundation for Statistical Computing, Vienna, Austria). Significant
150 differences were determined with the Kruskal-Wallis test ($\alpha = 0.05$) and Dunn's multiple comparison test
151 ($\alpha = 0.05$). Plots were achieved with *ggplot2*.

152 **Results and discussions**

153 **Evaluation of trace element contamination of Paris soils sampled in street tree pits**

154 TE concentration measurements in soils revealed a wide range of concentrations depending on the trees age,
155 location, and the TE considered (Fig. 2). Zn and Pb were the most abundant TE with a mean concentration of
156 $229 \pm 173 \text{ mg}\cdot\text{kg}^{-1}$ and $196 \pm 186 \text{ mg}\cdot\text{kg}^{-1}$, respectively. The mean Cd concentration was lower ($1.7 \pm 0.55 \text{ mg}\cdot\text{kg}^{-1}$)
157 than Pb and Zn concentrations, as frequently observed due to low Cd natural concentration in the environment
158 (Kabata-Pendias 2004; Azimi et al. 2005b). The heterogeneity of TE concentrations reflects the local influence
159 of road traffic density between streets. These concentrations were in line with results from soil samples covering
160 the whole Parisian region with a large range of anthropogenic pressures, but a limited number of samples from
161 Paris city (Gaspéri et al. 2016; Foti et al. 2017). Nonetheless, the concentrations measured in soils from Paris
162 were in the upper range of concentrations reported in other urban soils (ranging from 36 to $1641 \text{ mg}\cdot\text{kg}^{-1}$, 9 to
163 $252 \text{ mg}\cdot\text{kg}^{-1}$ and <0.2 to $2.45 \text{ mg}\cdot\text{kg}^{-1}$ for Zn, Pb and Cd, respectively; Table 2) and were included in the upper
164 limit concentration values allowed for sewage sludge application ($150\text{-}300 \text{ mg}\cdot\text{kg}^{-1}$, $50\text{-}300 \text{ mg}\cdot\text{kg}^{-1}$ and $1\text{-}3$
165 $\text{mg}\cdot\text{kg}^{-1}$ for Zn, Pb and Cd, respectively; European Directive 86/278/CEE).

166 To evaluate soil contamination, we calculated a pollution index (PI) defined as the ratio of the metal
167 concentration to the geochemical background concentration to evaluate the contamination of urban soils
168 compared to rural soils (Chen et al. 2005; Basioli et al. 2006). According to Basioli et al. (2006), $\text{PI} < 1$ reflects a
169 low contamination, $1 < \text{PI} < 5$, a moderate contamination, and $\text{PI} > 5$, a high contamination. However, in Paris,
170 street tree soils are imported from sites located in the vicinity of Paris ($<50 \text{ km}$) and placed in pits before
171 introduction of trees. Thus, we used data from Duigou and Baize (2010) on mean pedogeochemical background

172 in the region (estimated to $51.0 \pm 9.4 \text{ mg} \cdot \text{kg}^{-1}$, $23.1 \pm 8.3 \text{ mg} \cdot \text{kg}^{-1}$, and $0.3 \pm 0.1 \text{ mg} \cdot \text{kg}^{-1}$, for Zn, Pb, and Cd,
173 respectively) as geochemical background concentration. These values are close to those measured in rural soils
174 from the Parisian region with low anthropogenic influence (Saby et al. 2006; Foti et al. 2017). Thus, the PI
175 values we respectively calculated for Zn, Pb and Cd, i.e. 4, 8 and 5, indicated an important contamination of soils
176 in Paris.

177 TE emission related to traffic road, industrial activities, and residential heating was pointed out by many studies
178 as source of TE in atmospheric deposition in cities (Manta et al. 2002; Charlesworth et al. 2003; Basioli et al.
179 2006). In addition, atmospheric depositions strongly depend on the density of population of the city considered
180 (Charlesworth et al. 2003; Davis and Birch 2011). Since the anthropogenic origin of Zn, Pb and Cd has
181 frequently been reported in urban soils (Manta et al. 2002; Rodrigues et al. 2009; Ajmone-Marsan and Basioli
182 2010; Gaspéri et al. 2016) and since the street tree soils originally come from non-urban areas around Paris, the
183 TE concentrations very likely result from an anthropogenic origin linked to the urban environment such as urban
184 dust samples (de Miguel et al. 1997; Charlesworth et al. 2003; Ayrault et al. 2013). Moreover the high
185 population density in Paris might contribute to explain the important TE contamination we observed.

186 Many studies compared TE concentrations between soils of different cities and revealed important discrepancies
187 between cities probably due to differences in industrial activities and traffic road intensities (Madrid et al. 2006;
188 Rodrigues et al. 2009; Ajmone-Marsan and Basioli 2010). Azimi et al. (2005b) have estimated a total TE flux of
189 $103 \text{ mg} \cdot \text{m}^{-2} \cdot \text{y}^{-1}$ in Paris downtown, mainly originating from road traffic and residential heating. Zinc was the
190 most abundant anthropogenic metal measured in atmospheric depositions, representing around 50% of the total
191 measured TE (Zn, Pb, Cd and Cu), with constant level throughout the year, followed by Pb and Cu. However, Pb
192 isotope ratio calculated in aerosols from Paris indicated a shift in Pb sources since the 90's from a road traffic
193 origin to an industrial one (Widory et al. 2004). This shift was correlated with a decrease of TE deposition
194 (except for Cu and Zn) in the same period (Azimi et al. 2005a). The ban of leaded gasoline in 2000 could have
195 participated to this decrease, as already observed in Great-Britain urban areas (Charlesworth et al. 2003).

196 The time of residence of soil in Paris, i.e. tree age, significantly influenced soil TE concentrations (Fig.2). The
197 median concentrations exhibited the same pattern for Zn and Pb, with a statistically significant increase (Dunn
198 test, $p < 0.05$) from soils from young street tree pits to old street tree pits (i.e., for 65 years): from 62 to
199 $365.2 \text{ mg} \cdot \text{kg}^{-1}$ for Zn and from 47.36 to $307 \text{ mg} \cdot \text{kg}^{-1}$ for Pb, respectively, or six times the initial concentration.
200 Cd median concentration was close to $1.7 \text{ mg} \cdot \text{kg}^{-1}$, and no significant evolution of concentration with time was
201 evidenced. The increase of Zn and Pb in soils with the duration of the period spent in Paris underlined a TE

202 accumulation around $4.5 \text{ mg kg}^{-1} \text{ yr}^{-1}$ and $4 \text{ mg kg}^{-1} \text{ yr}^{-1}$ for Zn and Pb, respectively. Although accumulation
203 rates are rarely quantified, our results are consistent with results from other countries on street tree soils (Kagar et
204 al., 2013) and results comparing TE concentrations in park soils from different ages (Li et al. 2001; Madrid et al.
205 2002; Chen et al. 2005; Peltola et al. 2005; Madrid et al. 2006) or urban soils sampled twice with 25 years' time
206 interval (Imperato et al. 2003). Overall, our results show that it was very fruitful to use a soil chronosequence
207 based on street tree age to assess the mean long-term accumulation of TE. Such approach could be applied in all
208 towns where the soil of street trees is imported from outside towns and dumped each time a tree dies.

209 The PI drastically changed the evaluation of pollution for the different TE according to the age class considered.
210 Indeed, for Zn, young soils reveal a low pollution (PI = 1.2) (contrary to Pb and Cd, Zn being essential to plants,
211 in consequence PI of 1.2 could in consequence not be considered to be a pollution), medium soils a moderate
212 pollution (PI = 4.4), and old soils a high pollution (PI = 8.2), whereas Pb pollution is moderate in young soils (PI
213 = 2.05) and high in medium and old soils (PI = 8.4 and 16, respectively), despite Pb banning since 2000. These
214 differences of PI with age classes point to an accumulation of TE in soils mainly through airborne deposition and
215 street leaching (see above). The concentrations in soils from the old class were superior to the thresholds
216 recommended for sewage sludge application. No difference in soil Cd concentrations and PI between age classes
217 was evidenced. However, the PI indicated a high pollution (between 5 and 6.2) for the soils from the three age
218 classes. The high PI even for soil from the young age class suggests a quick accumulation in Cd in soils within
219 the first 15 years of street trees. The absence of Cd concentration increase in soils between younger and older
220 trees together with the fast contamination of the soils of the younger trees remained difficult to explain. A
221 possible explanation would be that there is a strong and recent source of Cd pollution. More generally, this points at
222 strong variations in Cd pollution within Paris during the last century.

223 **Trace element contamination of Paris trees**

224 Soil characteristics could influence TE bioavailability to plants (Kabata-Pendias 2004). However, in our study all
225 soils exhibited rather similar characteristics with a pH around 7.5, and a CEC between 11 and 13 cmol kg^{-1}
226 (Table 1). Thus TE bioavailability was likely equivalent, for example whatever the tree age, for each TE
227 considered. This suggests that differences observed between TE concentrations in tree biomass mainly reflected
228 changes in TE concentrations in soils rather than changes in the proportion of TE that is available. In addition,
229 atmospheric deposition should also contribute to tree contamination.

230 **Trace elements in roots**

231 Median concentrations of the different TE measured in roots were highly contrasted. Zinc was the most abundant
232 TE ($186 \text{ mg}\cdot\text{kg}^{-1}$), followed by Pb ($37 \text{ mg}\cdot\text{kg}^{-1}$) and Cd ($1.4 \text{ mg}\cdot\text{kg}^{-1}$). The high concentration in Zn in roots
233 might be due to the fact that Zn is an essential TE for plants contrary to Pb and Cd (DalCorso et al. 2014). Lead
234 values in roots were in the range of concentrations measured in linden roots from urban industrial sites in Serbia,
235 where soil Pb concentrations were slightly lower than in soils from Paris (Serbula et al. 2013). Zinc
236 concentrations in roots and soils from Paris were lower than concentrations in the Serbian sites. The TE
237 concentrations in roots, however, increased with tree age and the time spent in the city. Indeed, concentrations of
238 Zn and Pb in young tree roots were statistically lower than in old tree roots (Fig. 3). Despite low (*i.e.* below 1)
239 bioconcentration factors (corresponding to the ratio of TE concentrations in roots to TE concentration in soils),
240 TE concentrations in roots were statistically significantly higher in old soils than in young soils (Fig. 3). The
241 increasing TE concentration in roots with tree age is likely due to the progressive accumulation of Pb and Zn in
242 the soil and the transfer from soil to roots during the root life. Similar low bioconcentration factors from soil to
243 roots for Zn and Pb were evidenced by Serbula et al. (2013) in urban sites. This low bioconcentration reflecting a
244 low Pb transfer from soil to plant could result from i) its low bioavailability, which was often evidenced (Kabata-
245 Pendias, 2004), or ii) the age of the root sampled. Indeed, these root analyzed had a mean diameter $<2\text{mm}$ which
246 indicated that their residence time in soil was probably less than three years (Withington et al. 2006)

247 **Trace elements in leaves**

248 Tree leaves have been identified as useful biomonitors for TE deposition (reviewed by Gillooly et al. 2016), as
249 TE in the atmosphere can be trapped in the cuticular wax and trichome or even penetrate in stomata (Uzu et al.
250 2010; Schreck et al. 2012). However, in this study, leaf concentrations were measured to evaluate leaf
251 contaminations in TE according to tree age to evaluate a potential root (or soil)-leaf transfer, rather than for
252 biomonitoring.

253 TE median concentrations in leaves were low compared other cities: 0.01 , 0.8 , and $14.6 \text{ mg}\cdot\text{kg}^{-1}$ for Cd, Pb, and
254 Zn, respectively (Fig. 4) and more than ten-times lower than TE concentrations in roots. Similar leaf
255 concentrations were obtained from *Tilia* spp. leaves sampled in Belgrade by Anicic et al. (2011) and Deljanin et
256 al. (2016), whereas other studies have reported higher TE concentrations in *Tilia* spp. leaves from European
257 cities and from Istanbul (Piczak et al. 2003; Baycu et al. 2006; Sawidis et al. 2011; Schreck et al. 2012).
258 However a link between those higher leaf concentrations and a potential higher environmental contamination,
259 and transfer, to leaves could not be ascertained since these authors, did not measured TE concentrations in other

260 compartments (soil, root, or the atmosphere). In addition, leaves sampled in our study were washed. This could
261 have lowered the TE leaf concentrations measured as leaf washing removed around 10% of TE (deposited as
262 particulate on leaf surface) (Tomasevic et al. 2011; Deljanin et al. 2016). In particular, Cd and Zn can penetrate
263 into the leaf but Pb is mostly adsorbed to epicuticular lipids at leaf surfaces (Madejón et al. 2004).

264 The differences between leaf and root concentrations can be due to four complementary and non-exclusive
265 mechanisms: i) the fact that, for our target species, leaves are shorter-lived than roots and record an annual TE
266 signal whereas roots can grow in soil several years and can accumulate TE more than leaves, ii) a low root-leaf
267 transfer, iii) a low leaf contamination by direct airborne deposition and iv) a low foliar pathway transfer.

268 When considering soil-plant transfer, the low leaf TE concentrations measured in this study indicated no
269 statistically significant contamination of leaves despite different soil or root TE contamination. This suggests that
270 there was no significant transfer of TE from soils to leaves or from root to leaves. The low or negligible transfer
271 from soil to leaves was already noticed by Chojnacka et al. (2005) and Serbula et al. (2013) and could be due to
272 the speciation of these TE, with only a low proportion of TE being available for trees. For example, Ajmone-
273 Marsan and Biasioli (2010) indicated that in urban area Pb is adsorbed by Fe and Mn oxides and Pb exhausted
274 by vehicle is mainly present as particulate Pb (PbSO_4) with a very small proportion of Pb being soluble and thus
275 bioavailable for trees (Smith 1976; Harrison et al. 1981).

276 Thus, the contamination of leaves through airborne deposition appears to be the main likely contamination
277 pathway, explaining the low TE concentration in leaves. In addition, the main source of TE in Paris urban
278 environment reported was the atmospheric deposition (Rocher et al. 2004; Azimi et al. 2005b) and a
279 predominant foliar pathway for metal uptake compared to soil-root pathway for leafy plants (*e.g.*, lettuce,
280 parsley, and ryegrass) and pine was previously reported (Hovmand et al., 2009; Schreck et al. 2012), suggesting
281 that leaf contamination was mainly driven by airborne deposition. The transfer of Pb and Cd from airborne
282 sources to leaves was also observed by Gajbhiye et al. (2016a) at different sites of an industrial area, and for
283 roadside plants (Gajbhiye et al. 2016b). Moreover, some authors already recognize linden as valuable for
284 biomonitoring urban pollution (Sawidis et al., 2011; Serbula et al., 2013; Deljanin et al., 2016), especially for Pb.
285 In Paris, TE contamination was inferior to values reported in other European cities such as Venice (Rossini et al.
286 2005) or Belgrade (Mijić et al. 2010): 0.15 and 0.22 $\text{mg}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ for Cd, 3.6 and 21.7 $\text{mg}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ for Pb, and 29
287 and 41.4 $\text{mg}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ for Zn, respectively. And, according to data from Azimi et al. (2005), the atmospheric
288 deposition fluxes of TE in Paris have been decreasing between 1994 and 2002, reaching 29, 3.6 and 0.15

289 $\text{mg}\cdot\text{m}^{-2}\cdot\text{y}^{-1}$ for Zn, Pb and Cd, respectively, in 2002 as a consequence of the ban of Pb in fuel and an improved
290 treatment of flue gas. In consequence, the transfer of TE from the air to linden leaves might be low. The low
291 concentration in leaves could also indicate that the foliar pathway transfer for linden in this study was not
292 significant, despite a slight enrichment in Pb compared with linden leaves from a less urbanized area (data not
293 shown).

294 Finally, the atmospheric contamination of linden leaves in Paris could be compared with Belgrade city situation,
295 as atmospheric fluxes and TE leaf concentration are available (Mijić et al. 2010; Deljanin et al. 2016). TE
296 concentrations in linden leaves were in the same range between Paris and Belgrade, except for Pb, whose
297 concentrations were two times higher in Paris (Table 3). In Paris, Pb deposition fluxes were 6 times lower than in
298 Belgrade (Motelay-Massei et al. 2005). However, in Belgrade, leaded gasoline was still widely used, which
299 could explain the higher Pb fluxes in this city. The lower Pb concentrations in leaves from Belgrade than in
300 leaves from Paris could result from the localization of trees: in Paris, trees were in the vicinity of streets whereas,
301 in Belgrade, sampled trees were in a botanical garden, *i.e.* further from any street. Another factor might be
302 influential: the sampled linden trees may not belong to the same subspecies or even species in the two towns
303 studied, as the studies in Belgrade include a mix between *Tilia tomentosa* L. and *Tilia cordata* Mill. Both are
304 *Tilia* spp. (*e.g.* Aničić et al. 2011). This comparison reflected the importance of providing details on species and
305 localization of trees in the different sites or cities, as it likely strongly influences the results and their
306 interpretation. Especially, considering street or park trees, or considering trees of different ages for
307 biomonitoring can potentially lead to distinct results in terms of contamination. In consequence, this also
308 illustrates the difficulty of the application of biomonitoring to compare different sites. TE pollution
309 biomonitoring by city plants is frequently applied to evaluate the environmental quality or the impact of
310 industrial activities. However, as noticed by Mertens et al. (2005), biomonitoring of TE in plants presents some
311 drawbacks and the analysis of soils is also recommended.

312 In the actual context of increasing interest on urban agriculture, this study underlines the influence of TE
313 accumulation with time for soils exposed to urban environment and thus the necessity to evaluate soil
314 contamination before conversion of urban soil for urban agriculture. Moreover, when soil is imported from
315 outside towns to settle an urban farm, TE likely accumulate in this soil at rates that could be comparable to the
316 rates we assessed in street tree soils, but could depend on the proximity to streets, buildings and industries.
317 Nonetheless, the speciation and especially the bioavailability of the different TE in soils should be studied in
318 order to understand the fate of TE in urban soils and the possibility of using urban soils for agriculture on the

319 long term without contamination of the produced food. TE deposition on leaves is also of importance in urban
320 context and should be taken into account when the recycling of urban leaf litter is considered.

321

322 **Conclusions**

323 Our results indicate a pollution of soils for the three TE measured (Zn, Pb, and Cd). In addition, the increasing
324 soil concentrations in Zn and Pb from the young to the old class demonstrate an accumulation of TE with time.
325 This accumulation leads to concentrations higher than the usually recommended threshold values for sewage
326 sludge application, which questions the long term use of urban soils for urban agriculture. Although this increase
327 was not observed for Cd, the PI calculated for this element was consistent with an important pollution whatever
328 the age of the sampled soils.

329 However, tree roots indicated a low bioconcentration factor, despite a slight increase of TE concentration in the
330 old class roots. Thus, Zn, Cd, and Pb available fractions in these urban soils are supposed to be limited,
331 explaining the low soil-plant transfer. As the calculated bioconcentration factors from soil to leaves, and from
332 roots to leaves, indicated no significant transfer, leaf contamination should be mainly indicative of pollution
333 through airborne deposition.

334

335 **Acknowledgements**

336 We would like to acknowledge Yannick Agnan for comments on an early version of the manuscript. Sampling
337 campaigns benefited from funding from the Île-de-France region (R2DS), the GIS "Climate, Environment,
338 Society" (CCTV2 Project), the PIR IngEcoTech (IESUM project) and the Sorbonne Universities (Dens' project,
339 Convergences program).

340

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506 16
- 507

508

509 **Table 1:** Main soil characteristics of the three age classes of soil pits from Paris (France)

510

		Organic carbon content (%)	Total nitrogen content (%)	Clay (g kg ⁻¹)	Silt (g kg ⁻¹)	Sand (g kg ⁻¹)	CaCO ₃ (g kg ⁻¹)	CEC (cmol kg ⁻¹)
young class	Median	1.0	0.10	202	553	222	14	13.6
	Standard deviation	0.4	0.03	51	138	163	42	3.2
medium class	Median	1.7	0.14	165	299	489	49	12.5
	Standard deviation	1.1	0.05	41	89	115	53	3.8
old class	Median	2.0	0.18	100	115	672	112	11.2
	Standard deviation	1.2	0.08	41	140	153	31	2.8

511

512 **Table 2:** Mean trace element (Zn, Pb, Cd) concentrations of some urban soils (mg·kg⁻¹)

513

City	Country	Zn (mg·kg ⁻¹)	Pb (mg·kg ⁻¹)	Cd (mg·kg ⁻¹)	Reference
Palermo	Italy	138	202	0.68	(Manta et al. 2002)
Parisian region	France	210.4	102.8	0.4	(Gaspéri et al. 2016)
urban Parisian region	France	106-174	99-188	0.43-2.45	(Foti et al. 2017)
Paris	France	229	196	1.7	this study
Mexico	Mexico	36-1641	9-452		(Morton-Bermea et al. 2009)
Aveiro	Portugal	46	20		(Madrid et al. 2006)
Glasgow	Great Britain	199	307		(Madrid et al. 2006)
Ljubljana	Slovenia	114	78		(Madrid et al. 2006)
Sevilla	Spain	107	107		(Madrid et al. 2006)
Torino	Italy	225	144		(Madrid et al. 2006)
Uppsala	Sweden	112	47		(Madrid et al. 2006)
Sevilla	Spain	145	137		(Madrid et al. 2006)
London	Great Britain	108	158	<0.2	(Kelly et al. 1996)
Hong Kong	China	168	93.4	2.18	(Li et al. 2001)
Montreal	Canada	262	357	1.08	(Kargar et al. 2013)
Beijing	China	87.6	66.2		(Chen et al. 2005)
Torino	Italy	183	149		(Basioli et al., 2006)
Berlin	Germany	129	76.6	0.35	(Birke and Rauch, 2000)
Moscow	Russia	104	22.3	0.39	(Ermakov, 2017)

514

515 **Table 3:** Comparison of TE concentration and fluxes between Paris (France) and Belgrade (Serbia).

516

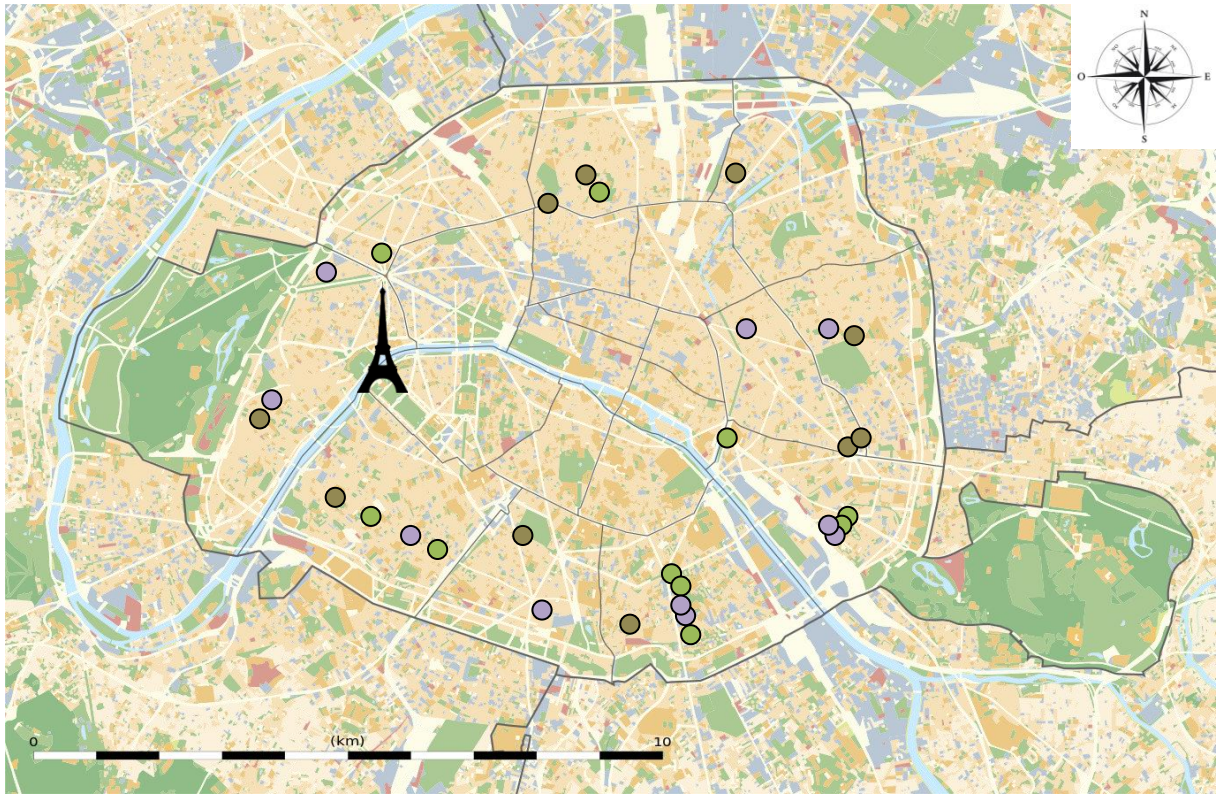
	City	Zn	Pb	Cd	References
Leaves ($\text{mg}\cdot\text{kg}^{-1}$)	Belgrade	10.00	0.50	0.02	(Deljanin et al. 2016)
	Paris	14.40	0.90	0.02	This study
Atmospheric fluxes ($\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$)	Belgrade	113.0	59.5	0.6	(Mijić et al. 2010)
	Paris	82.20	11.50	0.66	(Motelay-Massei et al. 2005)

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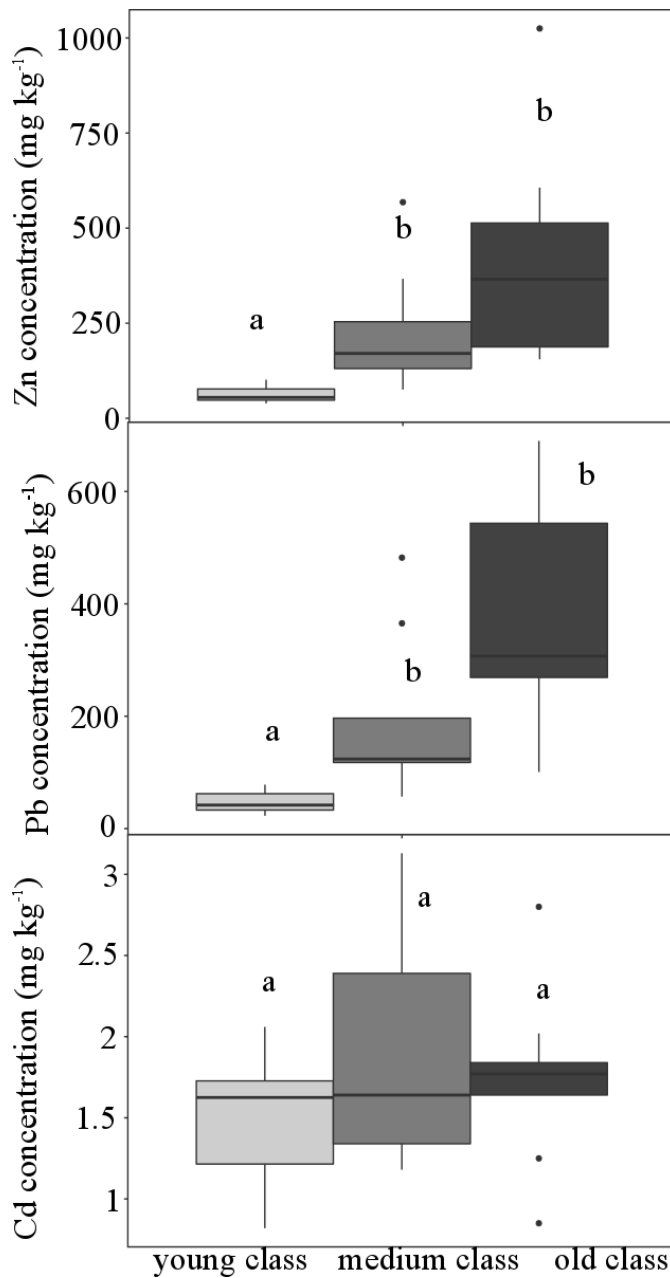


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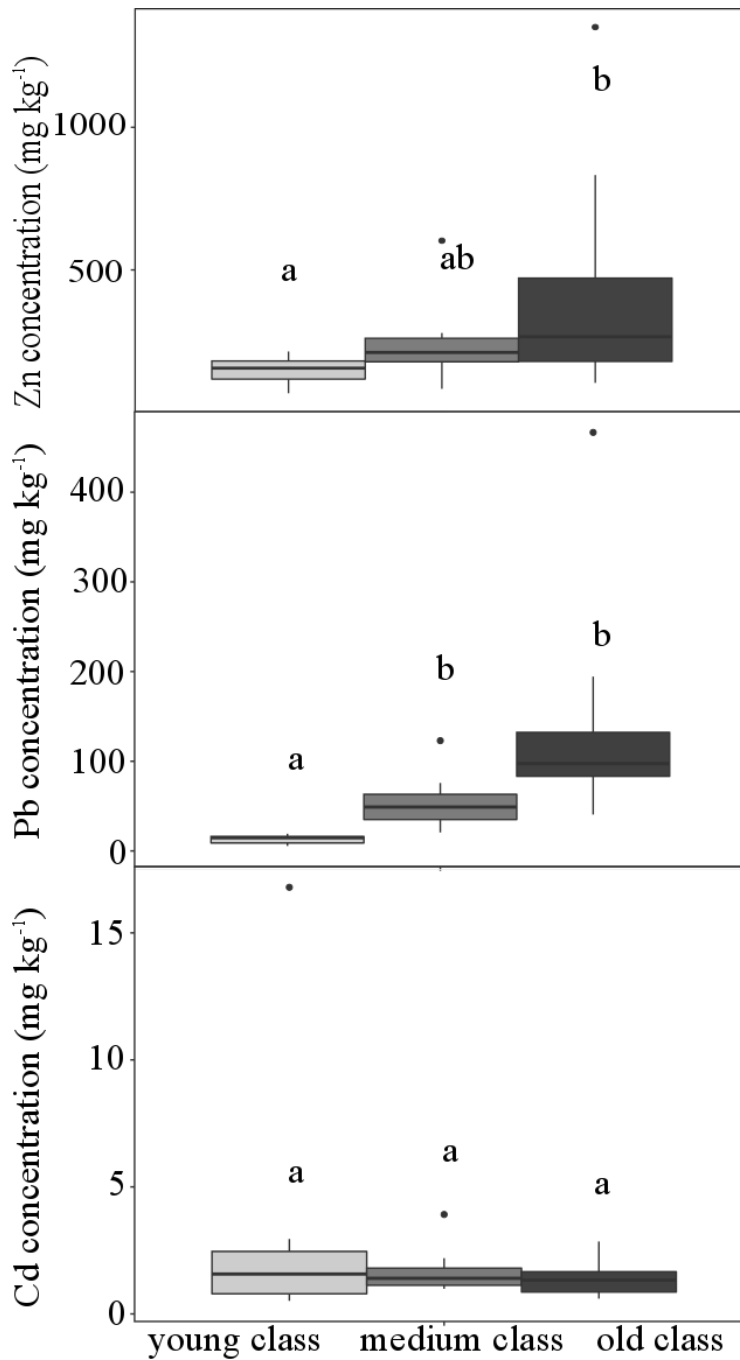
523 **Fig. 1** Localization of pit soils and trees sampled in Paris city, France (adapted from David et al. 2018). ● =
524 Young class; ● = Medium class; ● = Old class

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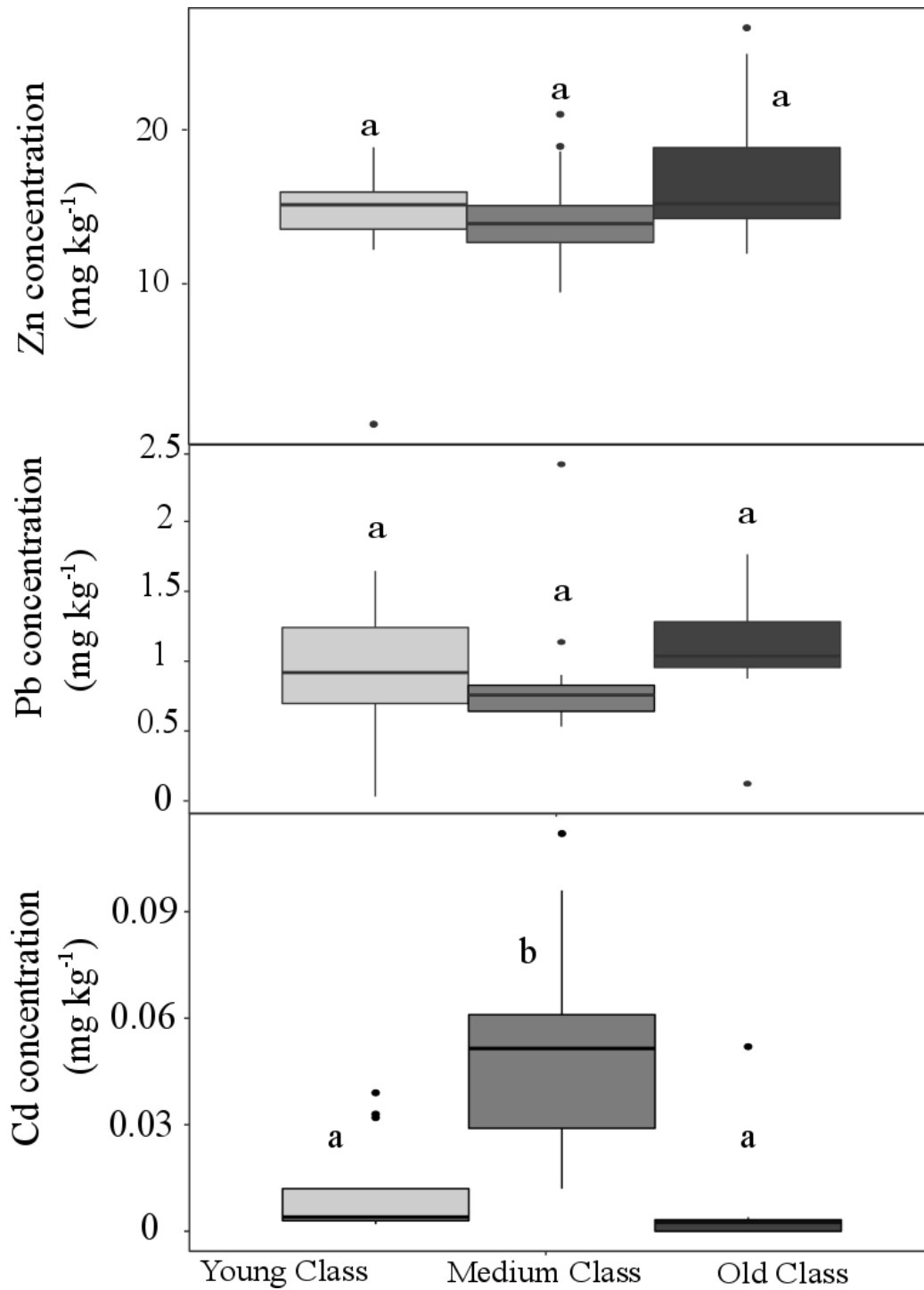
528 **Fig. 2:** Boxplots of Zn, Pb, and Cd concentrations in the soils of street trees of the three age classes (around 15
 529 years, 50 years and 80 years for young, medium and old class, respectively). Boxplot: horizontal bold lines of the
 530 box indicate the median, the lower and upper bounds of the box represent the 25th and 75th percentiles
 531 respectively. The vertical dotted bars include all values. Different letters indicate significant differences ($p < 0.05$)
 532 between the class ages (as determined by a Dunn test).



533

534 **Fig. 3:** Boxplots of Zn, Pb, and Cd concentration in the roots of street trees of the three age classes (centered
 535 around 15 years, 50 years and 80 years for young, medium and old class, respectively). Boxplot: horizontal bold
 536 lines of the box indicate the median, the lower and upper bounds of the box represent the 25th and 75th
 537 percentiles respectively. The vertical dotted bars include all values. Different letters indicate significant
 538 differences ($p < 0.05$) between the class ages (as determined by a Dunn test).

539



540

541 **Fig. 4:** Box plots of Zn, Pb, and Cd concentrations (mg kg⁻¹) in the leaves of street trees of the three age classes
 542 (around 15 years, 50 years and 80 years for young, medium and old class, respectively). Boxplot: horizontal bold
 543 lines of the box indicate the median, the lower and upper bounds of the box represent the 25th and 75th
 544 percentiles respectively. The vertical dotted bars include all values.

545