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RESEARCH ARTICLE

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 observed urban tree decline, while practitioners more and more question the possible reuse of these soils after the death of trees as well as tree litter collected in the streets. We evaluated the contamination in anthropogenic trace elements (TE), namely Zn, Pb, and Cd, of street trees (*Tilia tomentosa*) and their soils distributed all over Paris (France). Street tree soils are imported from rural areas at the plantation of each new tree so that tree age corresponds to the time of residence of the soil within an urban environment allowing the evaluation of temporal trends on TE concentration in soils and trees. The TE concentration revealed an important soil pollution, especially for the older soils (mean age of 80 years old). The consideration of the residence time of trees and soils in an urban environment evidenced an accumulation of Zn and Pb (ca. 4.5 mg kg^{-1} $year^{-1}$ and 4 mg kg^{-1} $year^{-1}$ for Zn and Pb, respectively). However, leaf concentrations in TE were low and indicate that soil-root transfer was not significant compared to the contamination by atmospheric deposition. These results underlined the necessity to deepen the evaluation of the recycling of urban soils or plants submitted to urban contamination.

Keywords Urban soils · Road traffic impact · Bioaccumulation · Trace element · Leaves · Roots

Introduction

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

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Many studies have focused on TE in urban environments either directly by soil concentration measurements (Ge et al. 2000; Manta et al. 2002; Maher et al. 2008) or through biomonitoring (Harmens et al. 2010; Deljanin et al. 2016; Gillooly et al. 2016). The use of vegetation as bioindicator of pollution is widely applied, plants being relevant integrator of their environment (Markert et al. 2003). Thus, measurements of TE concentrations in plants allow the assessment of the overall TE availability to plants without any distinction of the source (atmosphere, soil, water). However, all other things remaining equal (in particular, soil properties) TE concentrations in trees are influenced by tree species (Piczak et al. 2003; Pulford and Watson 2003), the precise localization of the street trees in the city (Maher et al. 2008), and the TE considered and its speciation (Kabata-Pendias 2004). Metals such as lead (Pb) and cadmium (Cd) accumulate close to intense traffic roads, and their concentrations decrease exponentially with increasing distance to the road (Viard et al. 2004; Nabulo et al. 2006; Werkenthin et al. 2014). Mosses and trees are frequently used in biomonitoring context (Markert et al. 1996; Piczak et al. 2003; Baycu et al. 2006; Gratani et al. 2008; Aničic et al. 2011; Sawidis et al. 2011; Guéguen et al. 2012; Natali et al. 2016). However, soil-plant transfer has often been neglected. In consequence, the simultaneous analysis of soil and plant present on a same site could help circumscribing limitations occurring when plants or soils are used separately for the evolution of site contamination (Mertens et al. 2005).

The city of Paris (France) is the fifth most important city in Europe in terms of population, with a high density (20,000 inhabitants/km^2). Human activities contribute to a large part of TE in Paris through industry emissions, vehicle exhaust, and residential heating. Despite a large decrease of TE deposition since 1994 (Azimi et al. 2005a), this atmospheric deposition remains the main source of TE, as evidenced by Garnaud et al. (1999) and Azimi et al. (2005a, b). However, another source of TE in urban soil is the runoff water from streets and roofs. In Paris, roof runoff was evidenced to be a significant source of Cd, Zn, and Pb (Garnaud et al. 1999; Gromaire et al. 2002; Rocher et al. 2004) and led to an increase of TE in the Seine River Basin (Thevenot et al. 2007). Nonetheless, extensive data on soil contamination by TE close to roads in Paris are still missing. The study of street trees growing in the vicinity of car exhaust and their soil could allow better assessing the extent of TE contamination in Paris. Indeed, atmospheric deposition, or street and roof leaching likely impact soils from those street trees and trees could be contaminated directly by either atmospheric deposition or by soil-plant transfer.

In Paris as in many cities, street trees are planted in pits that have been filed with agricultural soil transported from the country side (Paris Green Space and Environmental Division, pers. comm.). The soil is removed and replaced each time a tree dies and a young one is planted. This procedure provides a unique opportunity to assess the contamination dynamics through the comparison of trees with various ages (until 80 years) and their corresponding soil. Indeed, the age of street trees is also the residence time of the soil within the urban environment (Kargar et al. 2013). In addition, while TE accumulate during the whole tree life in their soils, TE accumulate in roots and leaves only during the life span of these organs. Thus, investigating root and leaf TE concentrations might allow distinguishing between the atmospheric or soil sources depending on the intensity of TE translocation between roots and leaves. Indeed, if translocation is low, root TE will mainly come from soil-plant contamination and leaf TE from atmosphere-plant contamination. In addition, leaves record the contamination during a single leafy season, whereas roots accumulate TE for several years depending on their size and life span (Withington et al. 2006). In addition, practitioners are so far dumping the soil of dead trees with the belief that these soils are no longer fertile, partially due to contamination by different sources of pollution. The same thing goes for street tree litter collected as it is considered as waste in the EU legislation (Nurmatov et al. 2016). Consequently, the evaluation of the soil and leaf contamination could help decide whether street tree soils and leaves should be reused or dumped after tree death and litter collection.

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

Materials and methods

Study area and sampling procedure

Thirty roadside linden trees (*Tilia tomentosa* Moench) were selected, as these trees are widespread in Paris (France) (48.8534° N; 2.3488° E). The selected roadside trees were gathered in three age classes: 10 trees belonged to the "young" class (between 11 and 17 years old, mean age of 13 years old), 10 trees to the "medium" class (between 41 and 67 years old, mean age of 49 years old), and 10 trees to the "old" class (69 and 86 years old, mean age of 80 years old). Tree age was determined by dendrochronological methods by David et al.

(2018). When a new tree is planted, a pit of about 3–4 m^3 is dug and filled with imported soil from surrounding peri-urban agricultural areas. Only vigorous trees with either bare or drain-covered soils were selected to avoid important differences in terms of rooting conditions and water availability (Rahman et al. 2011).

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

Sample treatment procedures

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

Pseudo-total metal concentrations were measured after soil mineralization. In the first step, soil samples were mineralized in aqua regia (mixture of 1/3 HNO_3 70% and 2/3 HCl 37%) using a temperature-controlled digestion system (DigiPREP Jr instrument, SCP Science, Baie-d'Urfé, Canada) at 120 °C for 8 h and dried.

Leaves and roots were mineralized according to the following procedure: Leaf and root samples were placed in Teflon flask with HNO_3 70% for 24 h at 120 °C with a DigiPREP instrument. After cooling H_2O_2, 30% was added and Teflon flasks placed at 120 °C for 24 h.

Physico-chemical analyses

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

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

$CaCO_3$ content, pH (H_2O), cation exchange capacity (CEC; Metson method), and soil texture results were provided by a soil analysis laboratory (INRA, Arras, France) according to standardized French procedures (AFNOR NF ISO 10693, NF ISO 10390, NF X 31-130, NF X 31-107, respectively).

Zinc concentrations were measured by inductively coupled plasma-optical emission spectrometry ICP-OES (instrument JY2000), whereas Pb and Cd concentrations were measured by inductively coupled plasma-mass spectrometry ICP-MS (X

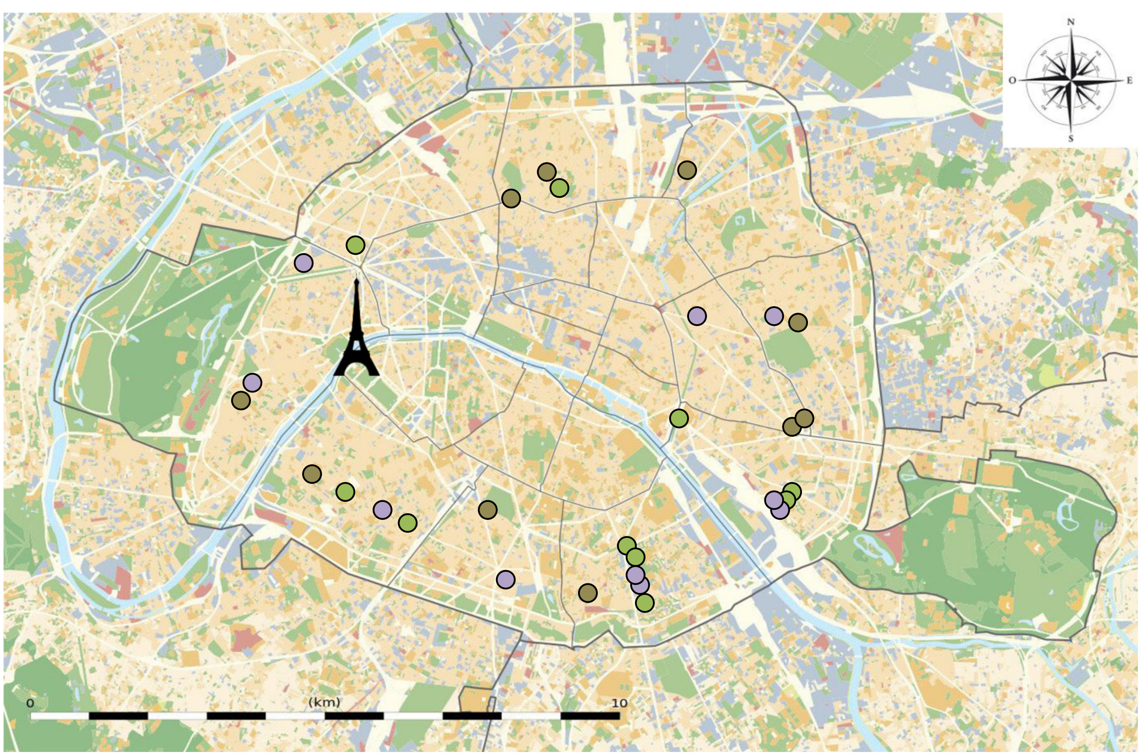

Fig. 1 Localization of pit soils and trees sampled in Paris City, France (adapted from David et al. 2018). ● = young class; ● = medium class; ● = old class

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

		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})	pH
Young class	Median	1.0	0.10	202	553	222	14	13.6	7.6
	Standard deviation	0.4	0.03	51	138	163	42	3.2	0.4
Medium class	Median	1.7	0.14	165	299	489	49	12.5	7.6
	Standard deviation	1.1	0.05	41	89	115	53	3.8	0.2
Old class	Median	2.0	0.18	100	115	672	112	11.2	7.5
	Standard deviation	1.2	0.08	41	140	153	31	2.8	0.2

Series II, Thermo Electron). Ten blank samples were added to the sequence following the same treatment for method control. Each sample was analyzed in triplicate. The detection limits of Pb, Cd, and Zn were 0.3, 0.2, and 2.2 μg L^{-1}, respectively, whereas the limits of quantification were about 0.4, 0.3, and 3 μg L^{-1}, respectively. The accuracy of calibrations was checked using a certified reference material (TMDA-64.2) from Environment Canada. The concentrations found corresponded to the certified values ± 5%.

Data treatment and statistical analysis

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

Results and discussions

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

TE concentration measurements in soils revealed a wide range of concentrations depending on the trees age, location, and the TE considered (Fig. 2). Zn and Pb were the most abundant TE with a mean concentration of 229 ± 173 mg kg^{-1} and 196 ± 186 mg kg^{-1}, respectively. The mean Cd concentration was lower (1.7 ± 0.55 mg·kg^{-1}) than Pb and Zn concentrations, as frequently observed due to low Cd natural concentration in the environment (Kabata-Pendias 2004; Azimi et al. 2005b). The heterogeneity of TE concentrations reflects the local influence of road traffic density between streets. These concentrations were in line with results from soil samples covering the whole Parisian region with a large range of anthropogenic pressures, but a limited number of samples from Paris City (Gaspéri et al. 2018; Foti et al. 2017). Nonetheless, the concentrations measured in soils from Paris were in the upper range of concentrations reported in other urban soils (ranging from 36 to 1641 mg kg^{-1}, 9 to 252 mg kg^{-1}, and < 0.2 to 2.45 mg kg^{-1} for Zn, Pb, and Cd, respectively; Table 2) and were included in the upper limit concentration values allowed for sewage sludge application (150–300 mg kg^{-1}, 50–300 mg kg^{-1}, and 1–3 mg kg^{-1} for Zn, Pb, and Cd, respectively; European Directive 86/278/CEE).

To evaluate soil contamination, we calculated a pollution index (PI) defined as the ratio of the metal concentration to the geochemical background concentration to evaluate the contamination of urban soils compared to rural soils (Chen et al. 2005; Basioli et al. 2006). According to Basioli et al. (2006), PI < 1 reflects a low contamination, 1 < PI < 5, a moderate contamination, and PI > 5, a high contamination. However, in Paris, street tree soils are imported from sites located in the vicinity of Paris (< 50 km) and placed in pits before introduction of trees. Thus, we used data from Duigou and Baize (2010) on mean pedogeochemical background in the region (estimated to 51.0 ± 9.4 mg kg^{-1}, 23.1 ± 8.3 mg kg^{-1}, and 0.3 ± 0.1 mg kg^{-1}, for Zn, Pb, and Cd, respectively) as geochemical background concentration. These values are close to those measured in rural soils from the Parisian region with low anthropogenic influence (Saby et al. 2006; Foti et al. 2017). Thus, the PI values we respectively calculated for Zn, Pb, and Cd, i.e., 4, 8, and 5, indicated an important contamination of soils in Paris.

TE emission related to traffic road, industrial activities, and residential heating was pointed out by many studies as source of TE in atmospheric deposition in cities (Manta et al. 2002; Charlesworth et al. 2003; Basioli et al. 2006). In addition, atmospheric depositions strongly depend on the density of population of the city considered (Charlesworth et al. 2003; Davis and Birch 2011). Since the anthropogenic origin of Zn, Pb, and Cd has frequently been reported in urban soils (Manta et al. 2002; Rodrigues et al. 2009; Ajmone-Marsan and Biasioli 2010; Gaspéri et al. 2018) and since the street tree soils originally come from non-urban areas around Paris, the TE concentrations very likely result from an anthropogenic origin linked to the urban environment such as urban dust samples (De Miguel et al. 1997; Charlesworth et al. 2003; Ayrault et al. 2013). Moreover, the high population density in

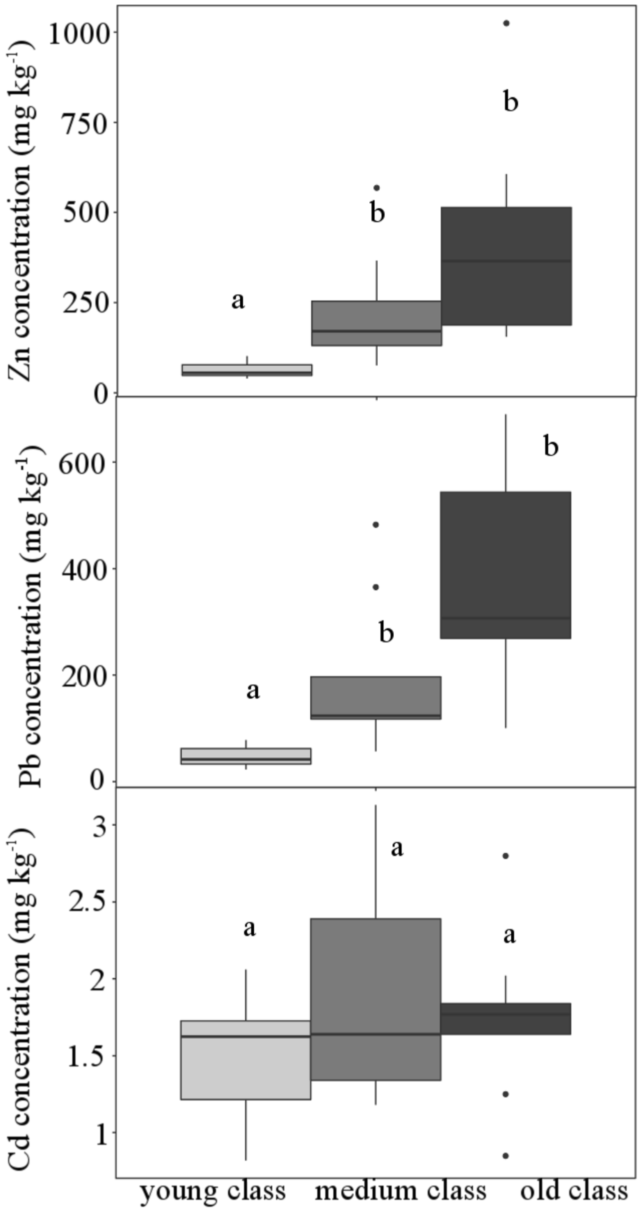

Fig. 2 Boxplots of Zn, Pb, and Cd concentrations in the soils of street trees of the three age classes (around 15 years, 50 years, and 80 years for young, medium, and old class, respectively). Boxplot: horizontal bold lines of the box indicate the median, and the lower and upper bounds of the box represent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

Paris might contribute to explain the important TE contamination we observed.

Many studies compared TE concentrations between soils of different cities and revealed important discrepancies between cities probably due to differences in industrial activities and traffic road intensities (Madrid et al. 2006; Rodrigues et al. 2009; Ajmone-Marsan and Biasioli 2010). Azimi et al. (2005b) have estimated a total TE flux of 103 mg m^{-2} year^{-1} in Paris downtown, mainly originating from road traffic and residential heating. Zinc was the most abundant anthropogenic metal measured in atmospheric depositions, representing around 50% of the total measured TE (Zn, Pb, Cd, and Cu), with constant level throughout the year, followed by Pb and Cu. However, Pb isotope ratio calculated in aerosols from Paris indicated a shift in Pb sources since the 1990s from a road traffic origin to an industrial one (Widory et al. 2004). This shift was correlated with a decrease of TE deposition (except for Cu and Zn) in the same period (Azimi et al. 2005a). The ban of leaded gasoline in 2000 could have participated to this decrease, as already observed in Great-Britain urban areas (Charlesworth et al. 2003).

The time of residence of soil in Paris, i.e., tree age, significantly influenced soil TE concentrations (Fig. 2). The median concentrations exhibited the same pattern for Zn and Pb, with a statistically significant increase (Dunn test, $p < 0.05$) from soils from young street tree pits to old street tree pits (i.e., for 65 years): from 62 to 365.2 mg kg^{-1} for Zn and from 47.36 to 307 mg kg^{-1} for Pb, respectively, or six times the initial concentration. Cd median concentration was close to 1.7 mg kg^{-1}, and no significant evolution of concentration with time was evidenced. The increase of Zn and Pb in soils with the duration of the period spent in Paris underlined a TE accumulation around 4.5 mg kg^{-1} year^{-1} and 4 mg kg^{-1} year^{-1} for Zn and Pb, respectively. Although accumulation rates are rarely quantified, our result are consistent with results from other countries on street tree soils (Kargar et al. 2013) and results comparing TE concentrations in park soils from different ages (Li et al. 2001; Madrid et al. 2002; Chen et al. 2005; Peltola et al. 2005; Madrid et al. 2006) or urban soils sampled twice with 25-year time interval (Imperato et al. 2003). Overall, our results show that that was very fruitful to use a soil chronosequence based on street tree age to assess the mean long-term accumulation of TE. Such approach could be applied in all towns where the soil of street trees is imported from outside towns and dumped each time a tree dies.

The PI drastically changed the evaluation of pollution for the different TE according to the age class considered. Indeed, for Zn, young soils reveal a low pollution (PI = 1.2) (contrary to Pb and Cd, Zn being essential to plants, in consequence PI of 1.2 could in consequence not considered to be a pollution), medium soils a moderate pollution (PI = 4.4), and old soils a high pollution (PI = 8.2), whereas Pb pollution is moderate in young soils (PI = 2.05) and high in medium and old soils (PI = 8.4 and 16, respectively), despite Pb banning since 2000. These differences of PI with age classes point to an accumulation of TE in soils mainly through airborne deposition and street leaching (see above). The concentrations in soils from the old class were superior to the thresholds recommended for sewage sludge application. No difference in soil Cd concentrations and PI between age classes was evidenced. However, the PI indicated a high pollution (between 5 and 6.2) for the soils from the three age classes. The high PI even

Table 2 Mean trace element (Zn, Pb, Cd) concentrations of some urban soils (mg kg^{-1})

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. 2018)
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 et al. 2017)

for soil from the young age class suggests a quick accumulation in Cd in soils within the first 15 years of street trees. The absence of Cd concentration increase in soils between younger and older trees together with the fast contamination of the soils of the younger trees remained difficult to explain. A possible explanation would be that there is a strong and recent source of Cd pollution. More generally, this points at strong variations in Cd pollution within Paris during the last century.

Trace element contamination of Paris trees

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

Trace elements in roots

Median concentrations of the different TE measured in roots were highly contrasted. Zinc was the most abundant TE (186 mg kg^{-1}), followed by Pb (37 mg kg^{-1}) and Cd (1.4 mg kg^{-1}). The high concentration in Zn in roots might be due to the fact that Zn is an essential TE for plants contrary to Pb and Cd (DalCorso et al. 2014). Lead values in roots were in the range of concentrations measured in linden roots from urban industrial sites in Serbia, where soil Pb concentrations were slightly lower than in soils from Paris (Serbula et al. 2013). Zinc concentrations in roots and soils from Paris were lower than concentrations in the Serbian sites. The TE concentrations in roots, however, increased with tree age and the time spent in the city. Indeed, concentrations of Zn and Pb in young tree roots were statistically lower than in old tree roots (Fig. 3). Despite low (i.e., below 1) bioconcentration factors (corresponding to the ratio of TE concentrations in roots to TE concentration in soils), TE concentrations in roots were statistically significantly higher in old soils than in young soils (Fig. 3). The increasing TE concentration in roots with tree age is likely due to the progressive accumulation of Pb and Zn in the soil and the transfer from soil to roots during the root life. Similar low bioconcentration factors from soil to roots for Zn and Pb were evidenced by Serbula et al. (2013) in urban sites. This low bioconcentration reflecting a low Pb transfer from soil to plant could result from (i) its low bioavailability, which was often evidenced (Kabata-Pendias 2004), or (ii) the age of the root sampled. Indeed, these

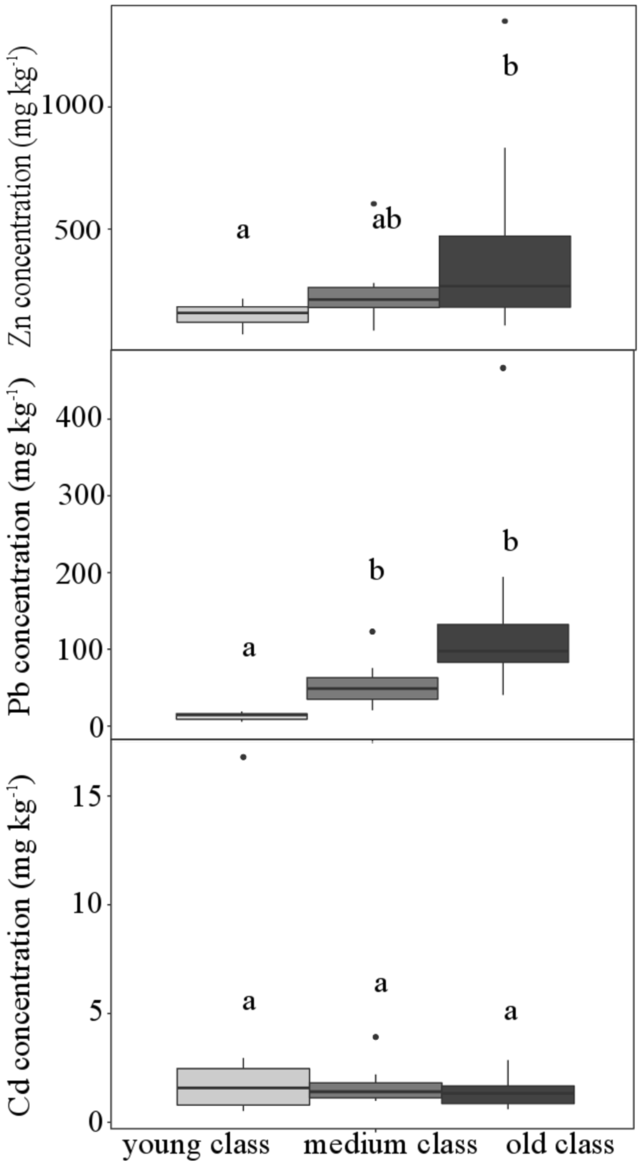

Fig. 3 Boxplots of Zn, Pb, and Cd concentration in the roots of street trees of the three age classes (centered around 15 years, 50 years, and 80 years for young, medium, and old class, respectively). Boxplot: horizontal bold lines of the box indicate the median, and the lower and upper bounds of the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

root analyzed had a mean diameter < 2 mm which indicated that their residence time in soil was probably less than 3 years (Withington et al. 2006).

Trace elements in leaves

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

TE median concentrations in leaves were low compared other cities: 0.01, 0.8, and 14.6 mg kg^{-1} for Cd, Pb, and Zn, respectively (Fig. 4) and more than 10 times lower than TE concentrations in roots. Similar leaf concentrations were obtained from *Tilia* spp. leaves sampled in Belgrade by Aničic et al. (2011) and Deljanin et al. (2016), whereas other studies have reported higher TE concentrations in *Tilia* spp. leaves from European cities and from Istanbul (Piczak et al. 2003; Baycu et al. 2006; Sawidis et al. 2011; Schreck et al. 2012). However, a link between those higher leaf concentrations and a potential higher environmental contamination, and transfer, to leaves could not be ascertained since these authors did not measure TE concentrations in other compartments (soil, root, or the atmosphere). In addition, leaves sampled in our study were washed. This could have lowered the TE leaf concentrations measured as leaf washing removed around 10% of TE

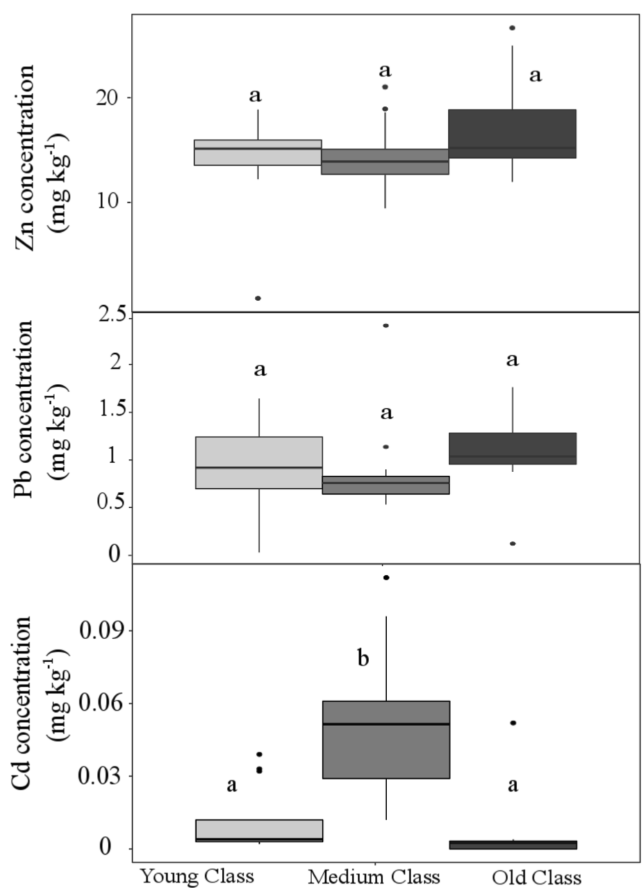

Fig. 4 Box plots of Zn, Pb, and Cd concentrations (mg kg^{-1}) in the leaves of street trees of the three age classes (around 15 years, 50 years, and 80 years for young, medium, and old class, respectively). Boxplot: horizontal bold lines of the box indicate the median, and the lower and upper bounds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

(deposited as particulate on leaf surface) (Tomašević et al. 2011; Deljanin et al. 2016). In particular, Cd and Zn can penetrate into the leaf, but Pb is mostly adsorbed to epicuticular lipids at leaf surfaces (Madejon et al. 2004).

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

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

Thus, the contamination of leaves through airborne deposition appears to be the main likely contamination pathway, explaining the low TE concentration in leaves. In addition, the main source of TE in Paris urban environment reported was the atmospheric deposition (Rocher et al. 2004; Azimi et al. 2005b) and a predominant foliar pathway for metal uptake compared to soil-root pathway for leafy plants (e.g., lettuce, parsley, and ryegrass), and pine was previously reported (Hovmand et al. 2009; Schreck et al. 2012), suggesting that leaf contamination was mainly driven by airborne deposition. The transfer of Pb and Cd from airborne sources to leaves was also observed by Gajbhiye et al. (2016a) at different sites of an industrial area, and for roadside plants (Gajbhiye et al. 2016b). Moreover, some authors already recognize linden as valuable for biomonitoring urban pollution (Sawidis et al. 2011; Serbula et al. 2013; Deljanin et al. 2016), especially for Pb. In Paris, TE contamination was inferior to values reported in other European cities such as Venice (Rossini et al. 2005) or Belgrade (Mijić et al. 2010): 0.15 and 0.22 mg m^{-2} $year^{-1}$ for Cd, 3.6 and 21.7 mg m^{-2} $year^{-1}$ for Pb, and 29 and 41.4 mg m^{-2} $year^{-1}$ for Zn, respectively. And, according to data from Azimi et al. (2005a, b), the atmospheric deposition fluxes of TE in Paris have been decreasing between 1994 and 2002, reaching 29, 3.6, and 0.15 mg m^{-2} $year^{-1}$ for Zn, Pb, and Cd, respectively, in 2002 as a consequence of the ban of Pb in fuel and an improved treatment of flue gas. In consequence, the transfer of TE from the air to linden leaves might be low. The low concentration in leaves could also indicate that the foliar pathway transfer for linden in this study was not significant, despite a slight enrichment in Pb compared with linden leaves from a less urbanized area (data not shown).

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

Table 3 Comparison of TE concentration and fluxes between Paris (France) and Belgrade (Serbia)

	City	Zn	Pb	Cd	References
Leaves (mg kg^{-1})	Belgrade	10.00	0.50	0.02	(Deljanin et al. 2016)
	Paris	14.40	0.90	0.02	This study
Atmospheric fluxes (µg m^{-2} 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)

environmental quality or the impact of industrial activities. However, as noticed by Mertens et al. (2005), biomonitoring of TE in plants presents some drawbacks and the analysis of soils is also recommended.

In the actual context of increasing interest on urban agriculture, this study underlines the influence of TE accumulation with time for soils exposed to urban environment and thus the necessity to evaluate soil contamination before conversion of urban soil for urban agriculture. Moreover, when soil is imported from outside towns to settle an urban farm, TE likely accumulate in this soil at rates that could be comparable to the rates we assessed in street tree soils, but could depend on the proximity to streets, buildings, and industries. Nonetheless, the speciation and especially the bioavailability of the different TE in soils should be studied in order to understand the fate of TE in urban soils and the possibility of using urban soils for agriculture on the long-term without contamination of the produced food. TE deposition on leaves is also of importance in urban context and should be taken into account when the recycling of urban leaf litter is considered.

Conclusions

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

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

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