Science of the Total Environment 598 (2017) 938–948

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Trace element concentrations along a gradient of urban pressure in forest and lawn soils of the Paris region (France)

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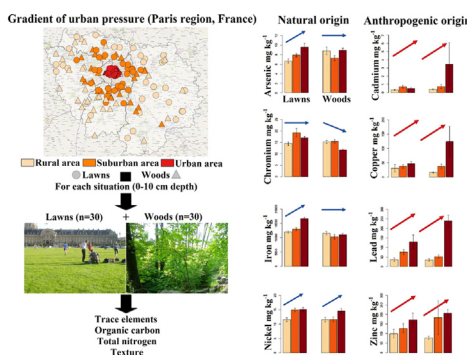
HIGHLIGHTS

- The anthropogenic trace elements are cadmium, copper, lead and zinc.
- The anthropogenic concentrations increase from the rural to the urban area.
- The first source of pollution for anthropogenic trace elements is the road traffic.
- Cement plants are the second source of cadmium.
- The trace element pollutions are impacted by the legacy of the soil history.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 17 February 2017
Received in revised form 13 April 2017
Accepted 14 April 2017
Available online xxxx

Editor: F.M. Tack

Keywords:
Trace elements
Urban-rural gradient
Soils
Green spaces
Lawns
Forests

ABSTRACT

The concentration, degree of contamination and pollution of 7 trace elements (TEs) along an urban pressure gradient were measured in 180 lawn and wood soils of the Paris region (France). Iron (Fe), a major element, was used as reference element. Copper (Cu), cadmium (Cd), lead (Pb) and zinc (Zn) were of anthropogenic origin, while arsenic (As), chromium (Cr) and nickel (Ni) were of natural origin. Road traffic was identified as the main source of anthropogenic TEs. In addition, the industrial activity of the Paris region, especially cement plants, was identified as secondary source of Cd. Soil characteristics (such as texture, organic carbon (OC) and total nitrogen (tot N) contents) tell the story of the soil origins and legacies along the urban pressure gradient and often can explain TE concentrations. The history of the land-use types was identified as a factor that allowed understanding the contamination and pollution by TEs. Urban wood soils were found to be more contaminated and polluted than urban lawns, probably because woods are much older than lawns and because of the legacy of the historical management of soils in the Paris region (Haussmann period). Lawn soils are similar to the fertile agricultural soils and relatively recently (mostly from the 1950s onwards) imported from the surrounding of Paris, so that they may be less influenced by urban conditions in terms of TE concentrations. Urban wood soils are heavily

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http://dx.doi.org/10.1016/j.scitotenv.2017.04.111

polluted by Cd, posing a high risk to the biological communities. The concentration of anthropogenic TEs increased from the rural to the urban areas, and the concentrations of most anthropogenic TEs in urban areas were equivalent to or above the regulatory reference values, raising the question of longer-term monitoring.

1. Introduction

Urban soils differ greatly from natural soils because they are located in areas of intense human activity, resulting in more pollution, physical disturbance and surface transformation. Whole soil profiles are often man-made, sometimes with fertile agricultural soils from rural areas (Bullock and Gregory, 1991; Beaudet-Vidal et al., 1998). Urban soils are responsible for a number of ecosystem services, including the recycling of organic matter and mineral nutrients and plant growth, and contribute to the ecosystem services provided by urban green spaces (from the mitigation of urban heat islands to recreational services) (Stroganova et al., 1997; Chiesura, 2004). Due to the potential impact urban soils can have on urban populations, there has been an increase in interest in urban soils in recent years (Wong et al., 2006; Morel and Heinrich, 2008; Vegter, 2007).

Urban soils can be polluted because they receive a variety of deposits from both proximal (vehicle emissions, domestic heating, waste incineration) and more distal (through atmospheric transport) sources (Islam et al., 2015). Trace elements (TEs) are persistent in the environment, tend to bioaccumulate in the food chain and, at high concentrations, can be toxic for humans and other organisms (Dudka and Miller, 1999; Raghunath et al., 1999; Adriano, 2001). When TEs accumulate in the soil, they impact the activities of soil organisms (microorganisms, microfauna, macrofauna), alter food web functioning, reduce the organic matter decomposition rate and disrupt biogeochemical cycling (Bååth, 1989; Alloway, 2013).

Some studies have documented urban soil TE pollution and its distribution at the scale of towns or their surrounding regions (Yesilonis et al., 2008; Amuno, 2013). In France, several monitoring programs (*e.g.* National Network for Long-term FOrest ECOsystem Monitoring (RENECOFOR), national soil quality monitoring network (RMQS) and studies have been carried out at a national scale, in order to measure the ranges of French soil TE concentrations and to estimate their natural background values in the country. While these studies brought significant information on French agricultural and forest soils (Baize and Sterckeman, 2001; Hernandez et al., 2003), only a few authors focused on urban soils (Joimel et al., 2016; Vergnes et al., 2017). This study aims at filling this knowledge gap, focusing on contamination and pollution by these TEs in lawns and forests, as they constitute the main types of vegetation in urban public areas of the region. Contamination occurs when a substance has a concentration above its natural background concentration while pollution is a contamination that results in adverse biological effects (Chapman, 2012; ISO Soil quality, 2005). Consequently, the degree of TE pollution in a soil describes the potential risks for the biological communities living in this soil and on its surface (*e.g.* microorganisms, plants). Contamination was evaluated for each single TE using the contamination factor (CF) and for all soil TEs together using the overall degree of contamination (DC). Pollution was evaluated for each single TE by the potential ecological risk index (ER) and by the ecological risk (RI) index for the overall pollution by all TEs (Hakanson, 1980; Amuno, 2013; Islam et al., 2015).

The aims of the present study were (i) to examine the concentration of seven selected TEs (As, Cd, Cr, Cu, Ni, Pb, Zn) and one major element (Fe) in soils from two land-use types (public lawns and woods) along an urban pressure gradient in the Paris region, (ii) to identify the origin of the diffuse and/or point sources of contamination or pollution, (iii) to evaluate the individual and overall TE contamination degree as well as the individual and overall TE pollution degree, (iv) to use soil characteristics to better understand soil origins and histories along the urban pressure gradient and the relationship between these characteristics and TE concentrations. The TEs analyzed were chosen because their concentrations are known to be influenced by human activities, and because it do not exist for these TEs a real monitoring of their concentration over time, especially in the soil of the urban public green spaces of the region (Paris Green Space and Environmental Division, pers. comm.). Ultimately, this study contributes to establish baseline TE values for the long-term monitoring of the evolution of TE soil contents in urban areas of the Paris region.

2. Materials and methods

2.1. Study area

The Paris region is located in France (48°07′N, 1°35′E; 49°07′N, 3°26′E) and covers an area of 12,070km^2 around the city of Paris. The population is 12.01 million, representing approximately 18.8% of the total population of metropolitan France (INSEE – French National Institute of Statistics and Economic Studies, 2013). The region is subject to several sources of anthropogenic trace elements (*e.g.* waste incineration, road traffic, metal smelter industries, Natali et al., 2016). The topography, geology and hydrology are relatively even across the whole region. It is characterized by an average altitude of 108m and very low erosion rates (10tkm^{-2}yr^{-1}) owing to limited relief. The bedrock is exclusively sedimentary (Jurassic limestone and marl, cretaceous chalk, carbonaceous alluvial deposits, tertiary quartz sand). The climate is subatlantic with an average temperature of 11°C and a rainfall of 600mm per year. The rainfall regime is pluvial oceanic (Pomerol and Feugueur, 1968). The climate is regionally marked by an urban and suburban heat island phenomenon (Tremeac et al., 2012).

2.2. Determination of urban pressure gradient of the Paris region

The urban pressure gradient of the Paris region was described using two spatialized indices, a Socio-Demographic Index (SDI) and a Heat Island Index (HII). The SDI is an average human activity density index per hectare of surface built. This type of index is frequently used in urban planning and territorial development (Frenkel and Ashkenazi, 2008). It allows the identification of the areas of a region that are most frequented by the population and that concentrate employment. The SDI map was made by the Paris Region Planning and Development Agency (IAU Île-de-France), by summing the population density and the employment density of the region (data set 2014, provided by INSEE). The result was normalized using the total built surface (data set 2014, provided by IGN – French National Geographic Institute). The spatial resolution of the SDI map was 250×250m. The HII uses the values of the minimal temperature recording to identify the areas that are most affected by human activities and the overall degree of artificial land cover (*e.g.* tar roads, buildings). The HII was calculated as the average of the monthly minimum temperature from January 2013 to December 2015 (data provided by Météo France – French Institute of Meteorological Prevision). The spatial resolution of the HII map was 2×2km.

To build the urban pressure map, the SDI and HII data were combined using GIS software (ArcGIS v.10) to obtain one map with a resolution of 2×2km. This was achieved by averaging the SDI values with an 8×8 filter to obtain a resolution of 2×2km. The urban pressure gradient was discretized into three classes (rural, suburban and urban areas) from a log-linear piecewise regression analysis of the SDI and HII datasets. No relationship between the SDI and the HII was observed for SDI below 10 inhabitant employment ha^{-1} and HII levels below 7.2°C (p-value=0.80), while a significant correlation was observed above these values (R^2=0.65, p-value<0.001). Three classes were considered in the analysis: a rural class for which the HII values are independent from the SDI values (<10 inhabitant employment ha^{-1} and <7.2°C, respectively), a suburban class (10–100 inhabitant employment ha^{-1} and 7.2–8.0°C, respectively) and an urban class (>100 inhabitant employment ha^{-1} and >8.0°C, respectively) for which the HII increases with the SDI (Fig. A).

2.3. Sampling design and protocol

180 sites were selected in public green spaces between September 28 and October 26, 2015. The sampling design followed a fully balanced cross factorial design with two factors: the gradient of urban pressure (UPG, 3 levels), and the land-use type (LT, 2 levels). The two land-use types targeted were lawns and woods, covering respectively 419 and 2670km^2 (3.5% and 22.2% of the region's area, ECOMOS 2003). In this study, woods are green spaces with a minimum continuous tree cover of 20ha. The woods selected for the study were mainly composed of deciduous species. All the combination of UPG and LT were equally sampled (n=30 by combination, n total=180) (Fig. 1, Appendix 1).

One composite soil sample was collected with an 8cm-diameter stainless steel hand auger at each site, immediately placed in polyethylene bags and subsequently air-dried in the laboratory. The composite sample was composed of 3 sub-samples collected within a 1m^2 square from the top 10cm of the organo-mineral soil layer and on flat ground. The composite samples were homogenized according to the NF X31-100 standard.

2.4. Trace and major elements and soil physico-chemical analysis

Seven TE (As: *arsenic*, Cd: *cadmium*, Cr: *chromium*, Cu: *copper*, Ni: *nickel*, Pb: *lead* and Zn: *zinc*) and one major element (Fe: *iron*) concentrations were measured following standard methods (Appendix 2). The samples were sieved to 2mm, followed by an automatic sieving conducted with an ultra-centrifugal mill<250μm (Retsch type MM400) to obtain a representative sub-sample (10g). Sub-samples were used for the TE and Fe determination after digestion with *aqua regia* (digestion block Labtech type ED36). The Cd, Cr, Cu, Ni, Pb and Zn concentrations were measured by direct Flame Atomic Absorption Spectrometry (F-AAS: Thermo Scientific ICE 3000 series). The concentration of As was measured by Hydride Generation Atomic Absorption Spectrometry (HG-AAS: Thermo Scientific IP/100). The concentration of Fe was measured by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES: Thermo Scientific ICAP 7000 series). HNO3, HCL (Chem-Lab) and double-distilled water were used for the preparation of samples, digestion solutions and standard solutions. Standard solutions of As, Cd, Cr, Cu, Fe, Ni, Pb and Zn (1000±2mgdm^{-3} in 5% HNO_3) were obtained from Chem-Lab. Standards for calibration were prepared daily by dilution from the standard solutions, and with the digestion solution. Calibration and blank samples were analyzed every 10 measurements. The accuracy of the determination of TE and major element concentrations was assessed using Internal Soil Reference Materials (ISRM1 and ISRM2 soils) for calibration using the same preparation procedures as the samples. The analytical precision, measured as the relative standard deviation, was below 2% in this study. There were 3 analytical replicates per sample. In order to minimize contamination from air, glassware and reagents, sample storage containers, such as centrifuge tubes, were rinsed and stored in a dilute acid bath (5% HNO_3) for several days prior to use. Teflon vessels and other lab ware were placed in a hot 20% HNO_3 bath for *circa* 4h and then rinsed several times with ultrapure water prior to use.

The main soil physico-chemical characteristics (silt, sand, clay, organic carbon (OC) and total nitrogen (tot N) soil contents) were measured following standard methods (Appendix 2). Soil texture was determined by the dispersion of mineral particles after destruction of the organic matter using hydrogen peroxide and separation of the particles into different classes by sedimentation. A dry combustion after sample burning at 1000°C in the presence of O_2 allowed quantification of the OC and the tot N soil contents (Waterlot et al., 2011).

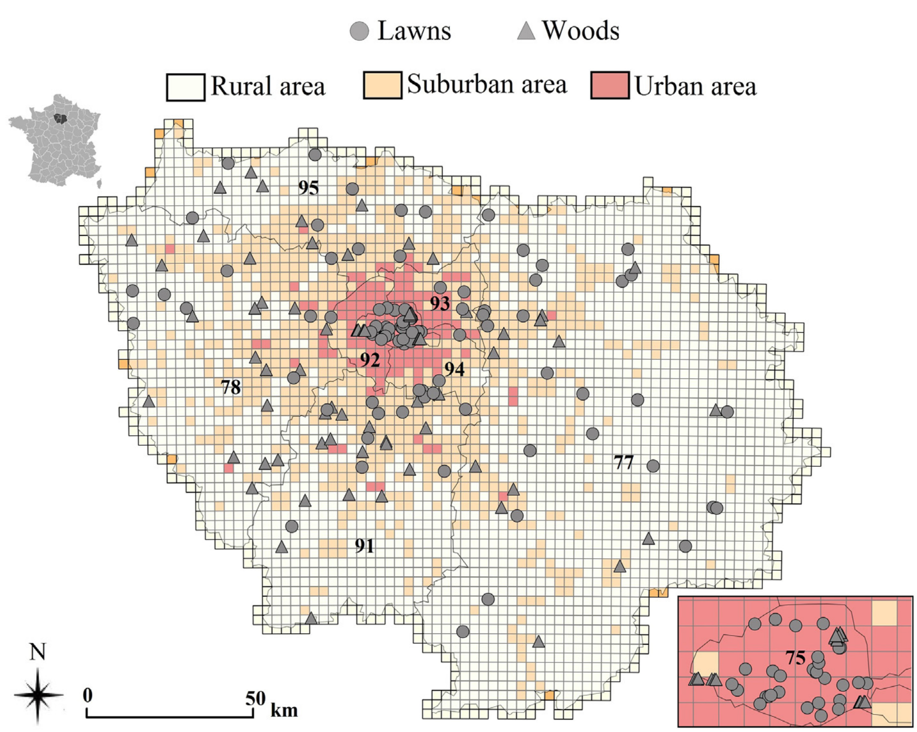

Fig. 1. Map (resolution: 2×2km) of the sampled sites (n=180) located from 48°07′N, 1°35′E to 49°07′N, 3°26′E with department delineation (75: Paris, 77: Seine et Marne, 78: Yvelines, 91: Essonne, 92: Hauts-de-Seine, 93: Seine-Saint-Denis, 94: Val de Marne, 95: Val d'Oise) in the Paris region (left) or in the urban area (right).

2.5. Data analyses

2.5.1. Trace element concentrations pretreatment

Some TE concentrations were below the limit of quantitation (LOQ, Appendix 2), *i.e.* below the minimum value that can be quantified using the method and apparatus used in this study. In order to make this data usable, when <25% of the values were below the LOQ value for a given TE, these values were transformed using the LOQ extrapolation method (Croghan and Egeghy, 2003). Initially selenium (Se) and mercury (Hg) concentrations were also measured but since >25% of the values were below the LOQ, these two TEs were not used in the statistical analyses.

2.5.2. Trace element origins and sources

A common way to distinguish anthropogenic from natural origins (*i.e.* the bedrocks) of TE is to estimate the enrichment factor (EF) with respect to local background TE values (assumed to be independent of any contamination by human activities). The background values published by Baize and Sterckeman (2001) were used as local background TE values. The determination of the EF requires the measured TE to be standardized against a reference element. In this study, iron (Fe) was taken as the reference element because its origin is believed to be strongly related to the bedrock (Reimann and de Caritat, 2005). The EF for each TE was calculated using the following equation: $EF_n=(C_n/B_n)/(C_{ref(n)}/B_{ref(n)})$, where C_n is the soil content of the TE of interest, $C_{ref(n)}$ the local background natural values of the TE of interest, B_n the soil content of the reference TE, $B_{ref(n)}$ the local background natural value of the reference TE (Table 1).

In order to determine whether TEs share common natural or anthropogenic origins and to identify potential sources of the contamination, a varimax rotation of the principal components analysis (PCA) was performed on centered and reduced TE data (Gallego et al., 2002). The varimax rotation is an orthogonal rotation that makes the interpretation of the factorial axes easier. In the case of TEs, the factorial axes obtained after the varimax rotation describe groups of TEs sharing the same origin. Finally, TEs associated with the factorial axis that is correlated to the reference element (Fe in the present study) are interpreted as having a lithogenic origin. The association of TEs with other axes indicates that they are of anthropogenic origin. Moreover, a TE associated with several axes point out several sources of contamination.

2.5.3. Trace elements contamination and pollution degree

The contamination factor (CF) accounts for the contamination of a single TE relative to its background reference value (Baize and Sterckeman, 2001). The sum of CF for all selected TEs represents the overall degree of contamination (DC) of the environment. The CF and DC formulae are given in Table 1. Individual TE ecological risk indexes (ER) were used to determine the degree of individual TE pollution. The ER formula (Table 1) used a specific biological toxic-response factor (TR) for each individual TE (10, 30, 2, 5, 6, 5, 1 for As, Cd, Cr, Cu, Ni, Pb and Zn, respectively). This study used the toxic response factors proposed by Hakanson (1980), who determined them in accordance with the toxicity level of each TE and the response of the biological communities to this toxicity level. The potential ecological risk (RI) is the sum of the ecological risk indexes (ER) for each TE. It is used as a diagnostic tool to determine the overall degree of TE pollution in soil (Qingjie et al., 2008). All TE index equations and grades are shown in Table 1.

2.5.4. Trace element concentrations, referential values and regulatory reference values

The TE level median concentrations of the Paris region were compared to referential values (RV) taken from the TE median concentrations of world soils and from 34 European city soils (Adriano, 2001; Yu et al., 2012). The TE concentrations of world soils represented typical TE contents in soils under variable conditions (*e.g.* bedrock type, soil texture) and including cultivated soils

Table 1
Equations for indices and grades of trace element enrichment, contamination and pollution factors. C_n: soil content of the investigated TE, $C_{ref(n)}$: natural background value of the investigated TE. B_n: reference TE value, $B_{ref(n)}$: natural background value of the reference element, TR: biological toxic factor of an individual TE.

	Indexes	Codes	Formula	Grades[a]	
Origin	Enrichment factor	EF	$EF_n=(C_n/B_n)/(C_{ref(n)}/B_{ref(n)})$	EF<2	Natural variability
				2≤EF<5	Moderate enrichment
				5≤EF<20	Significant enrichment
				20≤EF<40	Very high enrichment
				EF>40	Extremely highly enriched
Contamination	Contamination factor	CF	$CF_n=C_n/B_n$	CF<1	Low contamination
				1≤CF<3	Moderate contamination
				3≤CF<6	Considerable contamination
				CF≥6	Very high contamination
	Degree of contamination	DC	$DC_n=\Sigma CF_n$	DC<5	Low contamination
				5≤DC<10	Moderate contamination
				10≤DC<20	Considerable contamination
				DC≥20	Very high contamination
Pollution	Potential ecological risk index	ER	$ER_n=TR_n\times CF$	ER<40	Low risk
				40≤ER<80	Moderate risk
				80≤ER<160	Considerable risk
				160≤ER≥320	High risk
				ER≥320	Very high risk
	Potential ecological risk	RI	$RI_n=\Sigma ER_n$	RI<65	Low risk
				65≤RI<130	Moderate risk
				130≤RI<260	Considerable risk
				RI≥260	Very high risk

[a] The different grades were given according to Hakanson (1980) and Qingjie et al. (2008).

(possibly contaminated) and uncontaminated natural soils. The Regulatory Reference Values (RRV) are normative. They are generally based on background TE concentrations and on their toxicity level. In this study, the RRV were taken from the French association of normalization (on the median, AFNOR) (Baize, 1997) and from the European Community (on the range, Desaules, 2012). The French standard and European norm are actually used in the regulation of sludge spreading (Table 2).

2.5.5. Statistical analyses

A varimax rotation was performed with the psych R package (Revelle, 2016). The effects of the urban pressure gradient (UPG), the land-use type (LT) and their interaction (UPG×LT) were analyzed by linear models (Anova type I) with main soil physico-chemical characteristics, TE and major element (Fe: *iron*) concentration and enrichment factor (EF) as response variables. All variables were log transformed, except for Fe, in order to fulfill the hypotheses of the linear model. The proportion of the variance explained by each factor was calculated as the ratio between the sum of squares of each factor divided by the total sum of squares. Effects were compared using multiple comparisons of means (Tukey's Honestly Significant Difference). Spatial autocorrelation was evaluated for each linear analysis using the spdep R package (Bivand et al., 2012). A first model was fitted (response variable~factors) to check for spatial autocorrelation in the residuals using a Moran's I correlogram. The significance of Moran's I values was evaluated using a permutation test (n=1000). Since the autocorrelation was never significant, only models without the autocorrelation term were kept.

All statistical analyses were performed using the R software package (R Development Core Team, 2016).

3. Results

3.1. Soil physico-chemical characteristics

The UPG, the LT and UPG×LT had an impact on all soil physico-chemical characteristics (Table 3). The LT explained the most of the total variance of all the soil physico-chemical variables (13.4% on the average). It was followed by the part of the variance explained by the interaction effect (10.5% on the average).

Multiple mean comparisons showed that the effect of UPG×LT on soil physico-chemical characteristics was mainly due to the differences between the urban woods and the other woods and lawns in the three urban pressure categories. The organic carbon (OC), total nitrogen (tot N) and sand contents were highest in urban woods (55.6gkg^{-1}, 5.5gkg^{-1}, 68.4%, respectively), whereas the soil clay content showed opposite trends (10.1%). The pattern was more complicated for the silt content. The lowest silt content was found in urban woods (14.9%), the highest in rural lawns (42.2%), with intermediate contents in suburban and rural woods and suburban lawns (30.6%, 30.9% and 30.2%, respectively) (Fig. 2, Appendix 3). The lawn and wood soils were classified as Anthrosols (IUSS-WRB, 2014).

Table 3
Effect of the urban pressure gradient (UPG), land-use types (LT) and their interactions (UPG×LT) on the physico-chemical soil characteristics (OC: *organic carbon*, tot N: *total nitrogen*, Clay, Silt and Sand) (*p<0.05; **p<0.01; ***p<0.001). Results of linear models (degrees of freedom (df), F values, total residual df=174). (*p<0.05; **p<0.01; ***p<0.001.) All variables were log transformed.

	UPG	LT	UPG×LT	R^2
df	2	1	2	
OC	4.68*	29.52***	6.81**	0.23
tot N	5.63**	15.64***	10.09***	0.21
Clay	7.12**	19.38***	8.21***	0.22
Silt	8.29***	38.76***	12.06***	0.31
Sand	5.04**	13.60***	8.67***	0.19

3.2. Trace element concentrations

The UPG, the LT and UPG×LT impacted all TE and major element (Fe) concentrations but not for As, Cu and Zn concentrations, for which land-use type showed no effect (Table 4). The UPG explained a higher part of the variance (43.2% on average) than LT (11.61% on average) for all TEs, except Fe and Cr for which opposite trends were found. The variance of TE concentrations was more explained by LT than by UPG (LT: 15.44% on average; UPG: 8.26% on average). The interaction effect never explained the highest part of variance for Fe, and whatever the TE (11.44% on average).

The Cd, Cu, Pb and Zn concentrations were higher in urban woods than in all other woods and lawns (Fig. 3, Appendix 4). As, Cr, Fe and Ni showed opposite trends, with the highest As and Fe concentrations in urban lawns (9.6mgkg^{-1} and 16,594mgkg^{-1}, respectively), the highest Cr concentrations in suburban lawns (20mgkg^{-1}) and the highest Ni concentrations in urban and suburban lawns (115mgkg^{-1} and 115mgkg^{-1}, respectively) (Fig. 3, Appendix 4).

The patterns of Cu and Pb concentrations were similar, with a decreasing gradient in the following order: urban woods, urban lawns, suburban woods and lawns, rural woods and lawns (Fig. 3, Appendix 4). Cd and Zn concentrations followed a close pattern with the following decreasing gradient: urban woods, urban lawns, suburban woods and lawns, rural woods and lawns (Cd: 3.4, 0.5, 0.7, 0.7, 0.4 and 0.3mgkg^{-1}, respectively; Zn: 206, 171, 183, 127, 8 and 22mgkg^{-1}, respectively). Urban lawns had higher As concentrations (9.6mgkg^{-1}) than suburban woods and rural lawns (7.3 and 6.6mgkg^{-1}). There was no other significant difference between combinations of LT and UPG for As. Cr concentration was higher in suburban lawns (19.9mgkg^{-1}) than in urban

Table 2
Median trace and major element values (mgkg^{-1}) in lawns and woods from the rural, suburban and urban areas of the Paris region, referential values (RV) and regulatory reference values (RRV). (Trace elements=As: *arsenic*, Cd: *cadmium*, Cr: *chromium*, Cu: *copper*, Ni: *nickel*, Pb: *lead* and Zn: *zinc*; major element=Fe: *iron*.) nd=no data.

	Rural area		Suburban area		Urban area		RV		RRV	
	Lawns	Woods	Lawns	Woods	Lawns	Woods	34 European urban soils[a] *Median*	World soils[b] *Median*	AFNOR limit values[c] *Median*	European community limit values[d] *Range*
As	6.27	7.86	7.55	6.94	8.32	8.30	13.00	6.00	20	nd
Cd	0.28	0.30	0.67	0.65	0.43	2.45	0.95	0.35	2	1–3
Cr	15.05	15.06	15.70	15.68	16.47	11.97	59.00	70.00	150	nd
Cu	14.92	12.11	28.50	25.43	44.50	105.23	46.00	30.00	100	50–140
Fe	–		–		–		nd	nd	nd	nd
Ni	11.51	11.76	14.62	10.70	14.41	14.05	22.00	50.00	50	30–75
Pb	17.09	21.68	49.33	60.65	99.20	188.12	102.00	35.00	100	50–300
Zn	65.41	65.85	98.86	85.81	106.95	174.84	130.00	90.00	300	150–300

[a] Summarized in Yu et al., 2012, from 34 urban soils (Estonia, Germany, Greece, Ireland, Italy, Norway, Portugal, Serbia, Slovenia, Spain, Sweden and UK).
[b] Adriano, 2001.
[c] Sludge spread regulation (Baize, 1997).
[d] Compiled in Desaules, 2012 (from Bachmann et al., 1989), *section 8980* - for sludge amended soils.

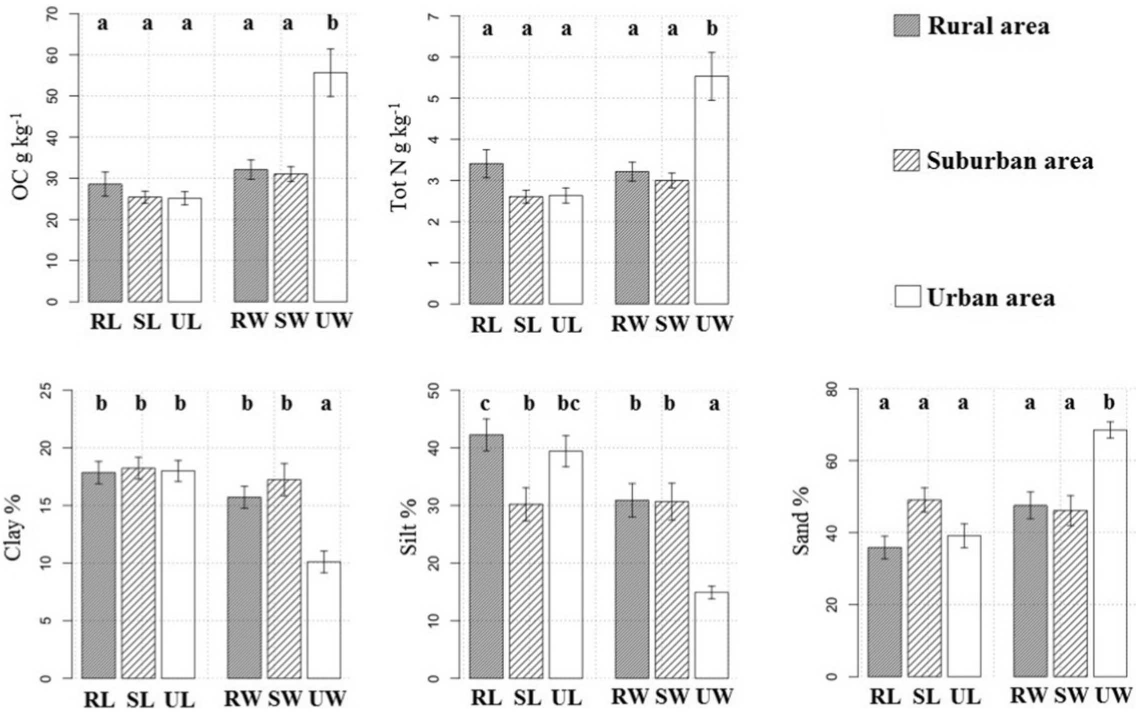

Fig. 2. Mean values of the physico-chemical soil characteristics in lawns and woods from the rural, suburban and urban areas of the Paris region. (OC: *organic carbon*, tot N: *total nitrogen*, Clay, Silt and Sand). RL: rural lawns, SL: suburban lawns, UL: urban lawns, RW: rural woods, SW: suburban woods, UW: urban woods. Error bars represent standard errors. Letters indicate significant differences between means.

woods (11.8mgkg^{-1}) and intermediate in all other kinds of sites. Fe showed a higher concentration in urban lawns (16,593mgkg^{-1}) than in all other sites. Its concentration was lower in suburban woods than in suburban lawns (10,254mgkg^{-1} and 12,925mgkg^{-1}, respectively). Ni showed lower concentrations in suburban woods and rural woods and lawns (11.6, 11.5 and 11.5mgkg^{-1}, respectively) than in urban and suburban lawns (15, 11 and 15mgkg^{-1}). Urban woods were not significantly different from any other type of site for Ni (Fig. 3, Appendix 4).

Table 4
Effect of the urban pressure gradient (UPG) and land-use types (LT) and their interactions (UPG×LT) on trace and major element concentrations and enrichment factors (EF) of the Paris region. (Trace elements=As, *arsenic*, Cd: *cadmium*, Cr: *chromium*, Cu: *copper*, Ni: *nickel*, Pb: *lead* and Zn: *zinc*; major element=Fe: *iron*.) Results of linear models (degrees of freedom (df), F values (F), residual df=174). (*p<0.05; **p<0.01; ***p<0.001.) All variables were log transformed, except for Fe concentration.

	UPG	LT	UPG×LT	R^2
df	2	1	2	
TE concentrations				
As	32.79***	0.5	3.62*	0.40
Cd	37.95***	14.04**	19.31***	0.61
Cr	8.07**	27.00***	12.14***	0.37
Cu	39.64***	0.34	17.44***	0.64
Fe	9.67***	40.65***	10.78***	0.31
Ni	60.91***	8.70*	15.66*	0.48
Pb	83.26***	14.89**	19.27***	0.66
Zn	83.97***	0.17	26.05***	0.62
Enrichment factors				
As	4.57	51.28***	16.42	0.07
Cd	78.43***	8.71**	17.42***	0.60
Cr	35.66***	53.83***	8.33***	0.50
Cu	43.88***	1.05	12.28***	0.64
Ni	35.40**	34.70***	86.30***	0.45
Pb	44.10***	7.03***	8.80***	0.64
Zn	43.97***	0.35	8.47***	0.60

3.3. Trace element origins and sources

The UPG, the LT and UPG×LT had an effect on TE enrichment factors (EF), except for Cu and Zn, for which land-use type had no effect, and for As for which UPG and UPG×LT had no effect (Table 4). UPG explained a higher proportion (48.85% on average) of the variance of EF than LT except for As and Cr, for which LT explained a higher proportion than UPG (28.07 and 27.96%, respectively), except for Ni for which UPG×LT explained the largest part of the variance (25.10%).

Higher EF were found in woods (0.97 on average) than in lawns (0.72 on average) for As. The Cr EF values were higher in suburban woods (0.81) than in urban lawns (0.48) with an intermediate EF value in urban and rural woods and in suburban and rural lawns (0.52, 0.63, 0.70 and 0.56, respectively). Ni showed lower EF values in urban and rural lawns (0.89 and 0.98) than in all other types of sites. Urban woods exhibited higher Ni EF values (1.3) than in rural woods (1.01) and in all type of lawns. All EF values for As, Cr and Ni belong to the natural variability range, indicating that they were from natural origin (Fig. 4, Appendix 5).

EF values of other TEs followed two different patterns. Cadmium and Zn EF values were higher in urban woods (on average 21.86 and 17.20, respectively) than in rural area (on average 1.87 and 3.00, respectively), with intermediate EF values in urban and suburban lawns and suburban woods (on average 4.39 and 1.61, respectively). Copper and Pb EF values decreased from urban woods (13.01 and 15.14, respectively), urban lawns (5.05 and 5.51, respectively) to suburban lawns and woods (on average 3.42 and 3.58, respectively) and rural lawns and woods (on average 1.62 and 1.84, respectively). For Cd, urban woods were very highly enriched, while for Zn they were significantly enriched. Suburban woods, for Cd and Zn, and urban and suburban lawns, for Zn, were also significantly enriched but urban and suburban lawns for Cd were moderately enriched. Urban area was significantly enriched for Cu and Pb, while suburban area was moderately enriched. Finally, rural area EF corresponded to the natural variability category for Cd, Cu, Pb, indicating that these TEs were from natural origin in soil of

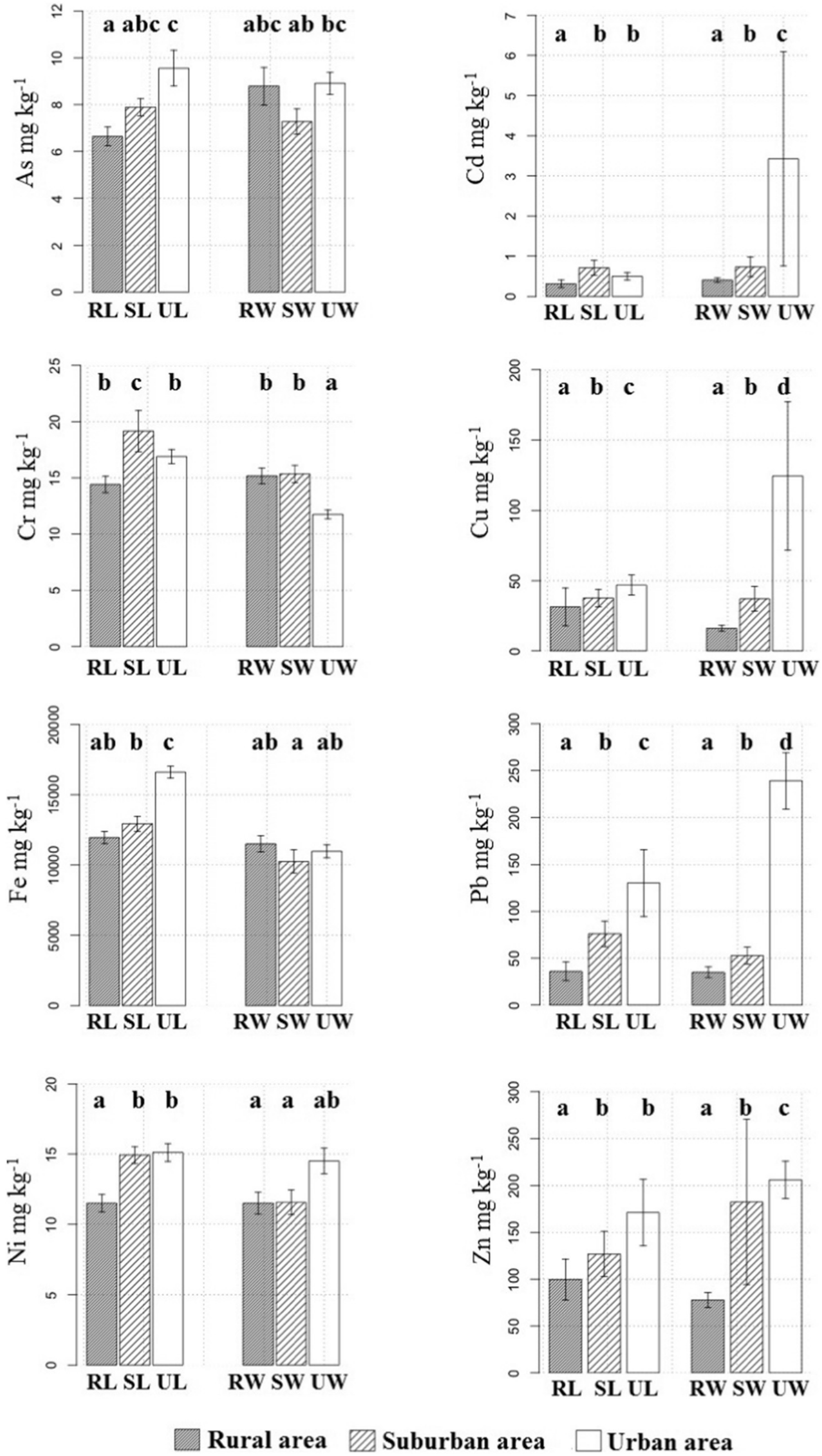

Fig. 3. Mean values of the trace and major element soil concentrations in lawns and woods in rural, suburban and urban areas of the Paris region. (Trace elements=As: *arsenic*, Cd: *cadmium*, Cr: *chrome*, Cu: *copper*, Ni: *nickel*, Pb: *lead* and Zn: *zinc*; major element=Fe: *iron*.) RL: rural lawns, SL: suburban lawns, UL: urban lawns, RW: rural woods, SW: suburban woods, UW: urban woods. Errors bars represent standard errors. Letters indicate significant differences between means.

rural woods and lawns unlike soil of woods and lawns in urban or suburban area. It should be noted that rural area was moderately enriched for Zn (Fig. 4, Appendix 5).

The first three rotated components of PCA on TE concentrations after the varimax rotation extracted 82% of total variance. Fe was correlated to the second axis (RC2), which identified this axis as representing natural sources of TEs and the others as representing anthropogenic sources. As, Cr and Ni were also correlated to the second axis, suggesting that they had natural origins and share a common lithogenic source, while Cd, Cu Pb and Zn were correlated to the first axis (RC1) suggesting that they had a common anthropogenic source. Lead, Zn and Cu were highly correlated with RC1 (0.88, 0.87 and 0.86, respectively), while Cd was moderately correlated with this axis (0.57) but also highly correlated (0.71) with the third axis (RC3). This indicated that Cd had two anthropogenic sources, one of which being the same as for Pb, Zn and Cu (Table 5).

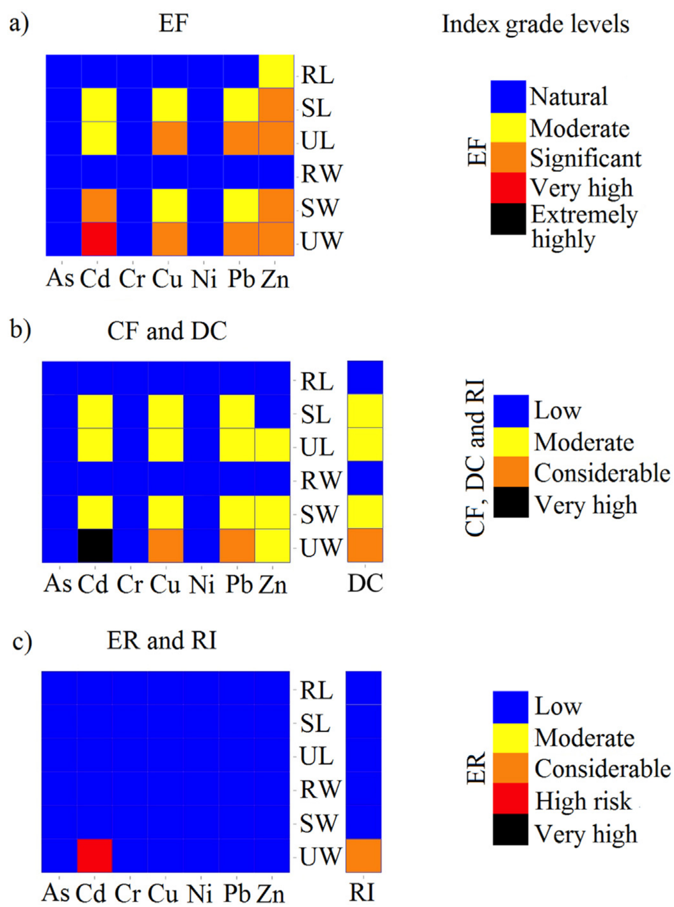

Fig. 4. Heat map of (a) trace element enrichment factors (EF), (b) individual trace element contamination factor (CF) and overall trace element degree of contamination (DC), and (c) trace element pollution degree through the individual trace element potential ecological risk index (ER) and the overall TE ecological risk (RI), in lawns and woods along the gradient of urban pressure. (As: *arsenic*, Cd: *cadmium*, Cr: *chrome*, Cu: *copper*, Ni: *nickel*, Pb: *lead* and Zn: *zinc*.) RL: rural lawns, SL: suburban lawns, UL: urban lawns, RW: rural woods, SW: suburban woods, UW: urban woods. The different grades were given according to Hakanson (1980) and Qingjie et al. (2008) (Table 1).

3.4. Individual, overall TE contamination and pollution degree

The response pattern for the Cd, Zn, Cu and Pb CF values were the same as for their EF values (Appendixes 5 and 6). According to the EF grades, urban woods were classified as very highly contaminated for Cd but moderately contaminated for Zn, while urban lawns and suburban areas were moderately contaminated for Cd and Zn. Urban woods were classified as considerably contaminated for Cu and Pb, while urban lawns and suburban lawns and woods were moderately contaminated. In addition, the rural lawns and woods were classified as lowly contaminated for Cd, Cu, Pb and Zn. All types of sites were classified as lowly contaminated for As, Cr and Ni. Finally, the overall degree of contamination (DC) was in the following order: urban woods (considerable contamination)>urban lawns=suburban woods=suburban lawns (moderate contamination)>rural woods=rural lawns (low contamination) (Fig. 4).

The response patterns for the ecological risk index values (ER) of Cd and Zn were the same as for their enrichment factors (EF) and contamination factors (CF). Similarly, Cu and Pb ER values responded in the same way as their EF and CF values. Finally, the response pattern for As, Cr and Ni ER values were the same as for CF (Appendixes 5 and 6). According to the ER grades, urban woods presented a high risk for Cd. All other sites, for all other TEs, presented a low risk. Finally, the ecological risk (RI) had the same response pattern as contamination factors (CF) (Appendixes 4 and 5). Urban woods presented a considerable ecological risk, while all other sites were considered as in low ecological risk (Fig. 4).

Table 5
Correlation coefficients of trace and major element concentrations with the first tree axes of the varimax PCA rotation after log transformation. The first lines provide the proportional eigenvalue for each axis. (Trace elements=As: *arsenic*, Cd: *cadmium*, Cr: *chromium*, Cu: *copper*, Ni: *nickel*, Pb: *lead* and Zn: *zinc*; major element=Fe: *iron*.) Correlation values≥0.5 are highlighted in bold.

Axis	RC1	RC2	RC3
Eigenvalues	39%	32%	11%
As	0.47	**0.65**	−0.43
Cd	**0.57**	0.01	**0.71**
Cr	−0.23	**0.83**	−0.26
Cu	**0.86**	0.25	0.17
Fe	0.08	**0.89**	−0.25
Ni	0.46	**0.75**	0.17
Pb	**0.88**	−0.06	−0.03
Zn	**0.87**	0.11	0.19

3.5. Trace element concentrations, referential values and regulatory reference values

The anthropogenic TE concentrations of Cd, Cu, Pb and Zn in both land-use types were generally higher than the world soil TE concentrations. The Cd concentrations were equivalent to the standard values of the French AFNOR regulation and to the European norms. Cu, Pb and Zn concentrations were lower than the European norm and the French AFNOR standard. All anthropogenic TE concentrations (Cd, Cu, Pb and Zn, as distinguished by the enrichment factors) of Paris region were higher than those found in European urban soils. In the urban area of the Paris region, all anthropogenic TEs were within the range of the European norm. The urban lawns showed Cd, Cu and Zn concentrations also equivalent to the ones found in European urban soils. For each anthropogenic TE, rural area showed lower concentrations than the world soil TE concentrations. Urban woods exhibited higher concentrations than the standard values of the French AFNOR regulation, except for Zn that had lower values. Finally, urban area showed Pb concentrations above all the referential values (Table 2).

The concentrations of natural origin TEs (As, Fe, Cr and Ni, as distinguished by the enrichment factors) presented systematically lower concentrations than the world soils, the European norm and the French AFNOR standard, except for the urban area for which the As concentration was relatively close to the world soils concentrations for both land-uses types (Table 2). It should be noted that a European norm has not yet been defined for As and Cr. It was the same for Fe, which is considered as soil major element (*i.e.* natural origin).

4. Discussion

4.1. Soil physico-chemical characteristics and soil origins

The soil physico-chemical characteristics of lawns were similar along the gradient of urban pressure. Moreover, the dates at which the urban and suburban public green spaces hosting the sampled lawns are relatively recent (1970 to 2011). This suggests that lawn soils could all have the same origin. Indeed, since 1950, arable lands have been artificialized in Paris conurbation area where Paris and its suburbs have been expanding. The fertile soils of these lands were excavated and resold as substrate for public green spaces of Paris conurbation (Paris Green Space and Environmental Division, pers. comm.), which is quite a common practice (Xie et al., 2005). Furthermore, the physico-chemical characteristics of these soils are constrained since 2004 by the NF U 44-551 standard (*e.g.* organic carbon, total nitrogen, clay, silt, sand), which could also partly explain the homogeneity of physico-chemical characteristics observed for lawns of the three categories of urban pressure. Note that the characteristics of the lawn soil (*e.g.* their texture) correspond to the properties of soils of croplands of the Paris region.

Unlike the lawns, the woods of the Paris region showed different physico-chemical characteristics along the urban pressure gradient. The urban woods were created during the period of Haussmann's renovation of Paris (1853 to 1867). Thus, the physico-chemical characteristics and the age of these urban woods strongly suggest a historical legacy of this period, when the urban public green spaces, such as urban woods, were profoundly recast (*e.g.* Vincennes, Boulogne) for creating market garden land (*e.g.* horticulture) or created (*e.g.* Buttes-Chaumont) to become recreation areas for the population (Gandy, 1999; Strohmayer, 2006). Market garden soils, that were relatively sandy, were used by Haussmann in public green spaces, because they were considered to be particularly fertile (Paris Green Space and Environmental Division, pers. comm.). In addition, during the Haussmann period, large quantities of wastewater were used for the irrigation of public green spaces and market garden land, because it was seen as a cheap mean of fertilization (due to high contents in organic carbon and total nitrogen) (Paris Green Space and Environmental Division, pers. comm.). This fertilization practice continued until 1950. Thus, the history of the soils likely explains the differences in physico-chemical characteristics observed between the urban woods and the other wood categories.

Furthermore, the differences in soil physico-chemical characteristics between woods and lawns (*i.e.* organic carbon and total nitrogen: Fig. 2, Appendix 3) could also be due to the effects of the two types of vegetation (*i.e.* high inputs of tree leaf litter and processes allowing the accumulation of a part of the carbon, Rout and Gupta, 1989; Blagodatskaya and Anderson, 1998). In comparison, the lawns of the Paris region are heavily managed, with several mowings per year after which the clipped grass is exported (Paris Green Space and Environmental Division, pers. comm.). This mowing regime could also partially explain that lawns and in particular urban lawns had lower organic carbon than urban woods (Hassink, 1994).

It is worth noticing that the statistical models only explained about 25% of the variability of the soil physico-chemical characteristics, which is low and much lower than for the variability of TE concentrations. This suggests that some important factors explaining the variability of the soil characteristics were missed. These characteristics might depend on soil natural heterogeneity for woods outside the urban area and on the precise origin of lawn soils, the origin of which has probably changed over time and between towns.

4.2. Trace element concentrations, origins and sources

The enrichment factors identified the existence of two TE groups in the Paris region. Woods presented different response patterns for anthropogenic TEs (Cd, Cu, Pb and Zn) and natural TEs (As, Cr and Ni), each of these natural TEs also having its own response pattern (Fig. B). For both land-use types, the anthropogenic TE concentrations and enrichments decreased along the urban pressure gradient, unlike the natural TE concentrations and enrichments. The anthropogenic TE concentrations and enrichments were mainly impacted by the urban pressure gradient and to a lesser extent by land-use type. Cd and Zn concentrations had very high standard deviations in urban and suburban woods, which suggests a heterogeneous spatial distribution for these TEs. This may be due to the intensification of soil manipulations (digging and soil exportation/importation) (Amuno, 2013).

In the Paris region, TEs with natural origin likely have lithogenic sources (Ajmone-Marsan and Biasioli, 2010), while Cd, Cu, Pb and Zn are known to have anthropogenic origins (Sternbeck et al., 2002). The Cu, Pb and Zn concentrations were highly correlated in the Paris region and are known as traffic markers (Stechmann and Dannecker, 1990). Cu and Pb are produced by brake emissions, Pb and Zn by tire wear emissions (Napier et al., 2008; Thorpe and Harrison, 2008) and Cd is also found in tires, diesel fuel and lubricating oils (Ajmone-Marsan and Biasioli, 2010). Despite the progressive replacement of leaded petrol by unleaded petrol since 1990 in France, and the definitive ban of Pb in 2000 (Flament et al., 2002), the Pb enrichment factor indicated a significant enrichment in urban areas and a moderate enrichment in suburban area. This could be explained by a long-term accumulation of Pb in roadside soils as Wong (1996) pointed out that all roadside soils contained more Pb than soils away from roadsides and their traffic. In the Paris region, densities of population and traffic volume are higher in urban area than in suburban area so that the higher anthropogenic TE enrichments in soils of urban area for both land-uses types must be the result of a high traffic road.

Cd likely has two anthropogenic sources. In addition to vehicle emissions, the industrial activities of the Paris region, especially cement plants that are authorized to use sewage sludge as an alternative source of fuel since 1992, are probably the second source of this TE. Rovira et al. (2014) observed increased concentrations of Cd in the local vegetation, following a switch to municipal sewage sludge as an alternative source of combustible by a cement plant.

Urban woods exhibited higher anthropogenic TE concentrations and enrichments, than all suburban and rural situations and urban lawns. As previously explained, urban wood soils, originated from market garden soils already rich in organic materials, were fertilized using wastewater as irrigation source in large quantities, beginning at Haussmann period and extending over more than a century. This wastewater not only contained organic matter but was also very rich in TEs (Paris Green Space and Environmental Division, pers. comm.). This may have impacted the urban woods TE concentrations and enrichments. As urban woods are older than urban lawns, they are more likely to have received wastewaters (Beaudet-Vidal et al., 1998; Paris Green Space and Environmental Division, pers. comm.). Moreover, their soils may have also accumulated more TEs than the soils from the other sites, especially for TEs produced by human activities (*e.g.* airborne inputs from traffic road, coal burning). This is supported by Chen et al. (2005), who showed that soil age is an important factor for TE accumulation in soils (*e.g.* Cu and Pb). Indeed, the older the soil, the longer the exposure to TE accumulation, and the higher the soil TE concentrations and enrichment factors. Finally, wood soils have higher organic matter contents than lawn soils. This could have contributed to the conservation and accumulation of TEs in urban woods as the soil organic matter (SOM) tends to adsorb TEs (Chen and Chen, 1996; Kabala and Szerszen, 2002).

4.3. Trace elements contamination, pollution degree, referential values and regulatory reference values, biological communities and human health risk

The main explication of individual and overall levels of TE contamination and risk (*i.e.* pollution level) is the urban pressure gradient especially for anthropogenic TEs. These results are in line with the findings of other studies on TE in urban areas (Li et al., 2001; Chen et al., 2005). Given the pattern of TE concentrations and enrichments along the urban pressure gradient, it was not surprising that individual and overall levels of contamination and risk are also the highest in the urban area. Despite these high levels of individual and overall contamination and risk, the associated grades of individual risk (ER) show that only urban woods present a considerable level of pollution. The Cd concentration being classified as leading to a “high” risk, and the associated overall risk (RI) leading to a “considerable” level of risk for biological communities (Fig. 4). This can be explained by the legacy of the historical management of soils in the Paris region and the higher age of urban wood soils (see above).

The TE concentrations of natural sources (As, Cr and Ni) were systematically below or equivalent to the referential values and below the regulatory reference values (world soils, European norm and French AFNOR standard, Adriano, 2001; Baize, 1997; Desaules, 2012, Table 2). The woods showed Cd concentrations equivalent to the French AFNOR regulation concentrations (Baize, 1997), the other anthropogenic TEs being below the regulatory reference values for both land-uses types. However, caution should be taken because the concentrations of anthropogenic TEs were high in urban and suburban areas and had higher concentrations than the standard values of the French AFNOR regulation (Baize, 1997), particularly in urban woods and especially for Cd, which could lead to toxic effect.

The toxic effects of TEs on soil micro- and macro-organisms are well-recognized (Bååth, 1989; Giller et al., 1998) even if contrasting trends have been reported on the effect of Cd on soil microbiota (Vig et al., 2003). The major factor governing the toxicity of a TE in soil is its bioavailability, which corresponds to the fraction of the total contaminant in the interstitial water and soil particles that is available to organisms (Naidu et al., 1997). Moreover, Cd shows more toxicity in sandy soils than in clay soils (Vig et al., 2003). Thus, the toxicity of Cd in urban woods of Paris is likely high because these woods combine high Cd and sand concentrations.

In addition to the potential risk to soil organisms, soil TEs could threaten human health, in particular for the level of Cd in urban wood soils (*e.g.* cancer risks, kidney tubular damage, Järup and Åkesson, 2009). Fine soil particles are known to have higher TE concentrations (Yu and Li, 2011). The fine particles of urban soils can be easily resuspended by airflows generated by wind or mechanical actions (*e.g.* traffic, human running). Hence, they are quite mobile in the environment and contribute significantly to the load of atmospheric particulate matters and total suspended particles, which is a major environmental concern in many cities (Young et al., 2002; Mossetti et al., 2005; Layton and Beamer, 2009). In addition, besides the inhalation of dust, urban soils could also be transferred to humans (especially children) through hand-to-mouth ingestion or dermal contact due to outdoor activities (Bright et al., 2006).

5. Conclusion

Determining the extent to which the overall pollution level we have described may adversely affect plants, animals, or even human health in the Paris region is beyond the scope of this paper. However, the high level of anthropogenic TEs found in the Paris area suggests that a long term monitoring program may be necessary. The concentrations of most anthropogenic TEs in urban area are either equivalent to or above the regulatory reference values. This likely arises from (1) the former use of wastewater for irrigation in parks, (2) human activities that lead to atmospheric deposition of TEs (mainly road traffic), (3) the use of sewage sludge as a combustible. Controlling TE concentrations thus requires controlling or reducing such activities. Urban woods require careful attention because: (1) they present high TE concentrations and risks for biological communities (*i.e.* pollution level), in particular for Cd; (2) they exhibit specific physico-chemical characteristics (high sand content, high organic matter content) that could strongly increase the TEs toxicity; (3) urban woods, such as the Bois de Vincennes and the Bois de Boulogne, are urban green spaces that are very frequented by the inhabitants of the region, especially by families and joggers. Besides, the development of urban agriculture in Paris region could increase the risks incurred by its inhabitants due to soil pollution by TEs. Indeed, plants have access to TE through their roots and vegetables grown in polluted soils can have high TE concentrations. In this context, our results confirm the idea that it is important to monitor TE in the soils of Paris region and follow movements of TE due to importation/exportation of soils. This suggests that urban agriculture in particular should be developed only in sites whose soil contamination level has been previously assessed.

Other studies are necessary to understand the dynamic of TE in soil of the Paris region to monitor where and which TEs can accumulate. This should involve studying: (1) all land-uses (and not only lawns and woods) including substrates that are made for green roofs and all forms of urban agriculture, (2) deeper soil layers and not only the surface soil layer, (3) factors involved in the dynamics of TE, *e.g.* factors determining the deposition and the loss of TE, (4) factors increasing the bioavailability of TE (*e.g.* chemical TE forms) and subsequent risks for humans.

Acknowledgements

This study was sponsored by the ANR ECOVILLE project (ANR-14-CE22-0021) and by NatureParif that has financed the PhD grant of the lead author. We would like to thank the public owners of the sampling sites for allowing the study to be carried out on their green spaces. We would like to thank the laboratory of agronomy of Paris, the laboratory of the analytical means of Dakar (IMAGO – IRD) and all associated technicians for providing the technical assistance in TE and soil physico-chemical analyses. We also would like to thank the IGN (France), the INSEE (France), Météo-France and the IAU (Île-de-France) for providing data.

Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.04.111.

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