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Heavy metal contamination of soil and tree-ring in urban forest around highway in Shanghai, China

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ABSTRACT

The heavy metal (HM) pollution of forest soil has been known as one of the most challenging pollution issues due to their characteristics. In order to know the HM pollution condition in urban forest, identify the possible source of HM, 102 sub-samples of soil in 34 sites and 39 tree rings sub-samples in 7 sites were collected in the outer-ring greenbelt (ORG) in Shanghai, China. Concentrations of Cu, Zn, Pb, and Cd in soil and tree rings were analyzed, and the soil properties pH, total nitrogen, total phosphorous, and organic matter were analyzed too. Geo-accumulation index and potential ecological risk index were used for assessing the contamination level of HMs. Nonparametric tests, one-way analysis of variance, correlation analysis, and principal component analysis were applied. The results showed that: (1) concentrations of Cu, Zn, and Cd in soil were significantly higher than their corresponding background values of Shanghai (BVs); concentrations of Cu, Zn, and Pb in tree rings increased gradually in the past 10 years; (2) Zn and Cd were in unpolluted to moderately polluted level, Cd has moderate degree potential ecological risk; (3) vehicle exhausts and abrasion of vehicle parts of tires and historical agricultural activities were the main sources of HM contamination; (4) *Cinnamomum camphora (L.) Presl.* has the potential to reconstruct the change of Cu, Zn, and Pb as a bioindicator. In conclusion, Cd should be considered as a priority control component. The relationship between plant and soil should take further focus and more studies of the behavior of HMs in soil and plants are required.

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greenbelt; soil; tree-ring; geo-accumulation index; potential ecological risk index

Introduction

Urban forests are an important part of the urban ecosystem, providing a myriad of ecological services that contribute and enhance human welfare, but at the same time, are profoundly influenced by urbanization (Ordóñez Barona 2015; Song *et al.* 2000). Due to rapid urbanization and industrialization during the past few decades, heavy metal (HM) concentrations in urban soils have reached a toxic level due to anthropogenic activities such as vehicle exhaust

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emissions, pesticide and fertilizer application, sewage sludge amendment, which release traces of HMs into the air, water, and soils (Liu *et al.* 2016; Peng *et al.* 2016; Sainger *et al.* 2011). The increasing accumulation and potential risk of exposure to HMs in urban soils have caused a growing public concern due to the potential harmful effects to human health, soils, and plants (Hsu *et al.* 2016; Zou *et al.* 2017). With the rapid urbanization and industrialization, China has become the world's leading heavy metal producer, resulting in soils contamination with HMs at different level (Hu *et al.* 2016; Mamut *et al.* 2017). The State Council of China issued the *Soil Pollution Control Action Plan* on May 28, 2016, put forward HMs such as Cd, As, Pb, Cr, and Hg as the key regulatory indicators (TSCC 2016), in order to strengthen soil pollution control and improve the soils' environmental quality.

Many studies investigated the accumulation and contamination of HMs, such as Cd, Cu, Pb, Cr, and Zn, particularly in farmlands and mining fields (Nriagu 1996; Barbieri *et al.* 2014; Larson 2014; Raj *et al.* 2017). Forest ecosystems that have been considered to be the most important terrestrial systems have been mostly overlooked and undervalued (Wingfield *et al.* 2015). The maintenance of forest ecosystems requires a functioning soil system, and the quality of forest soils will directly or indirectly affect the sustainable development of the forests (Castello and Teale 2011). Therefore, research focusing on the contamination and potential ecological risk levels of forest soil HMs is urgently required due to the significance of the soil in the forest ecosystem, especially in the urban forest ecosystem.

Previous studies linked changes in tree-ring growth and the environmental conditions. The use of tree rings as temporal monitors of environmental changes is possible because many tree species produce annual growth rings (Cocozza *et al.* 2016; Lepp 1975). Since Lepp reviewed the potential of tree rings chemistry for monitoring temporal changes in environmental trace metal levels (Lepp 1975), there has been a steadily increasing number of studies utilizing dendrochemistry for monitoring historical changes in trace metal levels in the soil, sediment, and atmosphere (Alahabadi *et al.* 2017; Panyushkina *et al.* 2016). In the last decades, many studies were carried out to reconstruct the temporal and spatial pattern by HMs recorded in tree rings of different species (Alahabadi *et al.* 2017; Watmough 1997). Therefore, in this study, concentrations of HM in tree rings were examined to reflect HMs temporal change and monitoring HM contamination of soil.

Shanghai has undergone rapid urbanization over the past several decades, its urbanization rate increased from 60.7% (1979) to more than 89.0% (2015) and became a commercial, economic, and industrial center for China. The population of the city was more than 24.1 million in 2015, and the population density is 3809/km^2 (SBS 2016). Meanwhile, increasing emissions of HMs related with a rapidly growing population and industrial activities have put enormous pressure on the environment and further exasperated ecosystem function. To improve environmental quality, the area of urban forest cover in Shanghai has been increasing annually, following afforestation efforts, its forest coverage increased from 3.0% (1979) to 14.04% (2015). Accompanied with rapid urbanization and increasing anthropogenic activities, and the demands for a better ecological environment, the levels of ecological risk exposure to HM in Shanghai are of concern. In the study described herein, the geo-accumulation index and potential ecological risk index were proposed to evaluate the contamination level of HMs in an urban forest in Shanghai, and the concentrations of HM in tree-ring were tested as a bioindicator of the soil HMs changeable. The aims of the study were to demonstrate the current status and historical variation of HM contamination in Shanghai's urban forest and to identify the possible sources of HMs.

Methods and materials

Study area

Shanghai is located on the coast of the East China Sea and at the estuary of the Yangtze River, which is a deltaic deposit of Yangtze River. The area of land region of Shanghai is about 6340 km^2, and the mean elevation is about 4 m above sea level. Except for several little volcanic massifs, the deposit was mostly formed during the Quaternary Era, and the modern soil mainly developed from later Holocene basement. The characteristic soil types are rice soil, gray alluvial soil, and coastal saline soil (Hou 1992; Xu *et al.* 2009; Zhang *et al.* 2015). The greenbelt around the Outer-Ring Highway in Shanghai was established in 1995, which was also the largest ecological project of green construction in Shanghai. After approximately 20 years of development, as the important part of urban forest, the ORG now has largely changed the ecological environment of Shanghai (Zhang *et al.* 2015). The length of the ORG is 98 km, encircling an area of 6208 hm^2, crossing seven city districts, which include Baoshan, Jiading, Putuo, Changning, Minhang, Xuhui, and Pudong. According to the establishment time, belt width, and community type, 34 sites in 13 sampling sections were chosen (Figure 1).

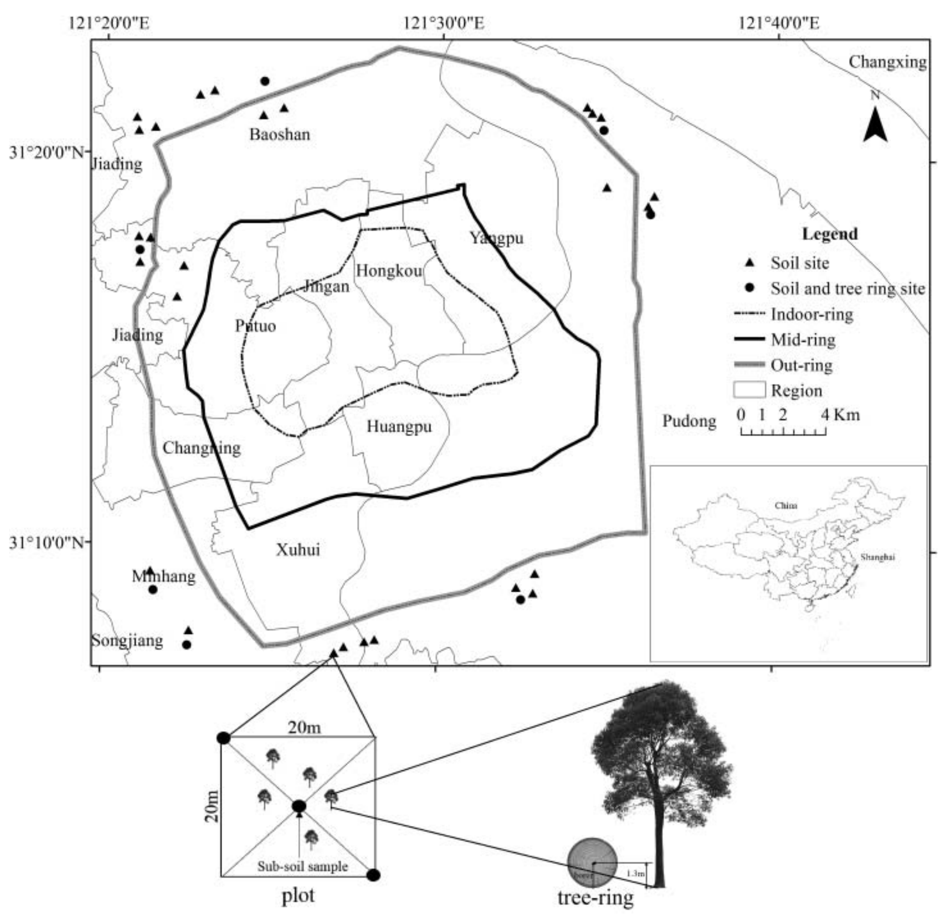

Figure 1. Location of sampling sites and schematic of samples collection (n = 34).

Sample collection and analysis

Sample collection

Soil sample collection was conducted in March and April of 2014. Each sample was a mixture of three sub-samples taken from the upper 0–30 cm of the topsoil. The sub-samples were excavated at 3 points on the diagonal line of 20 × 20 m plots in each location in accordance with the *Field sampling and preparation of forest soil samples of China* (CAF 1999). Soil samples were placed into clean polyethylene bags and transported to the laboratory, and then air-dried for 1 month, crushed by a pulverizer for 3 min (FW177, Pulverizer, China), and then sieved through a 0.15 mm nylon mesh for analysis.

The tree *Cinnamomum camphora (L.) Presl.* is an optimal choice for dendrochemistry in the region, consistent with the common ideals of wide range planted, native dominant (Zhang *et al.* 2011). Sampling and identification of this species are easy and inexpensive. Seven of the 34 study sites were chosen for tree rings analysis, in which four or five healthy *C. camphora* individuals were randomly collected and sampled. For each individual, two orthogonal tree cores were collected from the main trunk at a height of 1.3 m above the ground using 5.15 mm stainless steel manual increment borer(CO600, Haglof, Sweden) (Cocozza *et al.* 2016; Song *et al.* 2011). All tree cores were sealed in rigid plastic straws and dried naturally. The tree-ring width was measured using WinDendro tree-ring image analysis software (Version 2003a; Regent Instruments, Sainte-Foy, QC, Canada) to the nearest 0.001 mm. Two tree cores from each tree were cross-dated using the real-time cross-dating features of WinDendro, which facilitate the detection of false rings and missing rings (Song *et al.* 2011). Because the trees were transferred from other places at different ages, only the tree rings of the latest 10 (2004–2013) years were chosen for analysis. Because of small mass of ring growth, we divided the dataset into three periods after dendrochronological analysis to get better results: adaptation to the local environment period (2004–2008); steady tree growth period (2009–2012); and the last growth year (2013) (Panyushkina *et al.* 2016). Finally, the tree-ring fractions were crushed for 3 min (FW177, Pulverizer, China) and sieved through a 0.25 mm nylon mesh for analysis (Cocozza *et al.* 2016; Panyushkina *et al.* 2016).

Sample analysis

Four HMs were selected for this study (Cu, Zn, Pb, Cd), being the most common HMs in Shanghai and significantly harmful to human, plant, and animal health (Larson 2014; Wang *et al.* 2017). The soils' main properties: pH, total nitrogen (TN), total phosphorus (TP), and organic matter (OM) were measured, due to their fundamental effect on the concentration of HMs (Vega *et al.* 2004; Zhao *et al.* 2010).

Soil pH was measured by mixing soil with deionized water at a ratio of 1:2.5 (soil: water), and then the supernate was tested using glass electrode after standing for 1 h. Soil TN and TP were determined using an autodiscrete analyzer (Odyssey Clx, Clever Chem 200, Germany) and OM was determined with total organic carbon analyzer (Vario TOC, Elementar Analysensysteme GmbH, Germany).

For concentration of the HMs (Cu, Zn, Pb, Cd) in the soils, 0.25 g (precise to 0.0001 g) of each dry sample were digested for 3 h in a microwave oven, and the digestion was carried out with a concentrated acid mixture (5 mL HF, 10 mL HNO_3, and 2 mL $HClO_4$). Next, the samples were placed on an electric hot plate at 300°C till the solution were boiled, standing for 12 h, and then heated again at 300°C until complete dissolution of the soil. The total concentration of HMs was analyzed by atomic absorption spectroscopy (AANALYST 800 model, Perkin Elmer Company, USA). Pb and Cd concentration was measured using graphite furnace atomic absorption spectroscopy, and Cu and Zn concentrations were measured using flame-atomic absorption spectroscopy. The obtained sample solutions were diluted with deionized water to 50 mL and analyzed in triplicate, and total metal concentrations were calculated on a dry weight basis. The detection limit for Pb, Cd, Cu, and Pb was 0.78 μg/L, 0.039 μg/L, 0.016 mg/L, and 0.03 mg/L, respectively.

For tree-ring samples, 0.15 g (precise to 0.0001 g) each dry sample was digested in a microwave oven with a concentrated acid mixture (8 mL HNO_3, 2 mL $HCLO_4$). The same methods that were applied for the soil analysis were used to analyze the HMs concentrations in the tree-ring samples, but temperatures of 160°C and 230°C were used.

Assessment methods

Geo-accumulation analysis methods

The geo-accumulation index (I_{geo}) was originally used for sea-bottom sediments during the late 1960s (Muller 1969), and now it has been successfully applied to the measurement of soil contamination (Barbieri 2016; Ololade 2014). The index of I_{geo} enables the assessment of contamination by comparing with current and pre-industrial concentrations. It can be computed using the following equation:

$$I_{geo} = \log_2(C_n/1.5B_n) \tag{1}$$

where C_n is the measured concentration of the elements in soil and B_n is the geochemical reference value. We used the background values of the study area or the relevant landscape limited values provided by as the Chinese State Environmental Protection Administration geochemical reference survey (Table 1). The constant 1.5 is used to account for the possible variations in the reference values as they are affected by natural fluctuations and anthropogenic influence (Aiman *et al.* 2016). Muller's classification was used to assess the level of contamination (Table 1).

Potential ecological risk index

The I_{geo} can reflect the influence of concentration accumulation of a single HM and does not consider the bioavailability or combined effects of HMs. Therefore, the assessment of soil contamination was also carried out using the potential ecological risk index proposed by Hakanson (1980).

Table 1. Classification for the I_{geo}.

Value	Class	Soil quality
$I_{geo} \le 0$	0	Practically uncontaminated
$0 < I_{geo} \le 1$	1	Uncontaminated to moderately contaminated
$1 < I_{geo} \le 2$	2	Moderately contaminated
$2 < I_{geo} \le 3$	3	Moderately to heavily contaminated
$3 < I_{geo} \le 4$	4	Heavily contaminated
$4 < I_{geo} \le 5$	5	Heavily to extremely contaminated
$5 < I_{geo}$	6	Extremely contaminated

Table 2. Indices and grades of potential ecological risk.

E_r^i	Grades of ecological risk for a single metal	RI	Grades of potential ecological risk on the environment
$E_r^i < 40$	Low risk	$RI < 150$	Low risk
$40 \leq E_r^i < 80$	Moderate risk	$150 \leq RI < 300$	Moderate risk
$80 \leq E_r^i < 160$	Considerable risk	$300 \leq R < 600$	Considerable risk
$160 \leq E_r^i < 320$	High risk	$600 \leq RI$	High risk
$320 \leq E_r^i$	Very high risk		

It can be used to evaluate the potential ecological risk by soil HMs to humans, animals, and plants. This comprehensive method considers four factors: concentration, type of pollutant, toxicity level, and the sensitivity of the soil to HM contamination and illustrates the potential ecological risk caused by overall levels of contamination (Table 2) (Hakanson 1980). The equation used for calculation of potential ecological risk index is as follows:

$$RI = \sum E_r^i = \sum T_r^i \left(\frac{C_r^i}{C_n^i} \right) \tag{2}$$

where RI is the sum of individual potential ecological risks for all HMs. E_r^i (EI) is the potential ecological risk index of a single HM, T_r^i is the toxic-response factor for a given HM, C_r^i is the present concentrations of HMs in soils, and C_n^i is the reference values of HMs. In this study, we used the background values of Shanghai (BVs) (Wei 1990), and the grade II values of environment quality standard for soils in China (GBs) (SEPA 1995) as the reference values. The BVs for Cu, Zn, Pb, and Cd are 26.0, 79.9, 24.5, and 0.12 mg/kg. The GBs for Cu, Zn, Pb, and Cd is 200.0, 300.0, 350.0, and 0.6 mg/kg.

The toxic-response factors for Cu, Zn, Pb, and Cd are 5, 1, 5, and 30 (Hakanson 1980).

Statistical analysis

As data did not follow a normal distribution, nonparametric Kolmogorov–Smirnov tests were performed to identify the differences between HMs concentrations tested in this study and their related reference values. One-way analysis of variance (ANOVA) was applied to test the differences of HMs concentrations in tree rings between different periods. Pearson's correlation analysis was performed to assess the relationships between soils HM concentrations and soil properties, and relationships between HMs concentrations in tree rings and soil properties (SPSS ver. 23). Principal components analysis (PCA) was conducted to further assist the identification and analysis of sources of HMs in soil. To interpret the results more precisely, the rotation method-Varimax with Kaiser Normalization was applied to maximize variances of the squared normalized factor loadings across variables for each factor. All principal factors extracted from variables were those whose eigenvalues were higher than 1. Log-transformations were performed since these raw data sets did not follow a normal distribution pattern. The box-plot was conducted with software R, ver. 3.2.0.

Results and discussion

Soil HMs concentration

In this study, the GBs are outside of the range of the boxplots' axis; therefore, we only listed the BVs in the boxplots. Compared with the GBs, 200.0, 300.0, 350.0, and

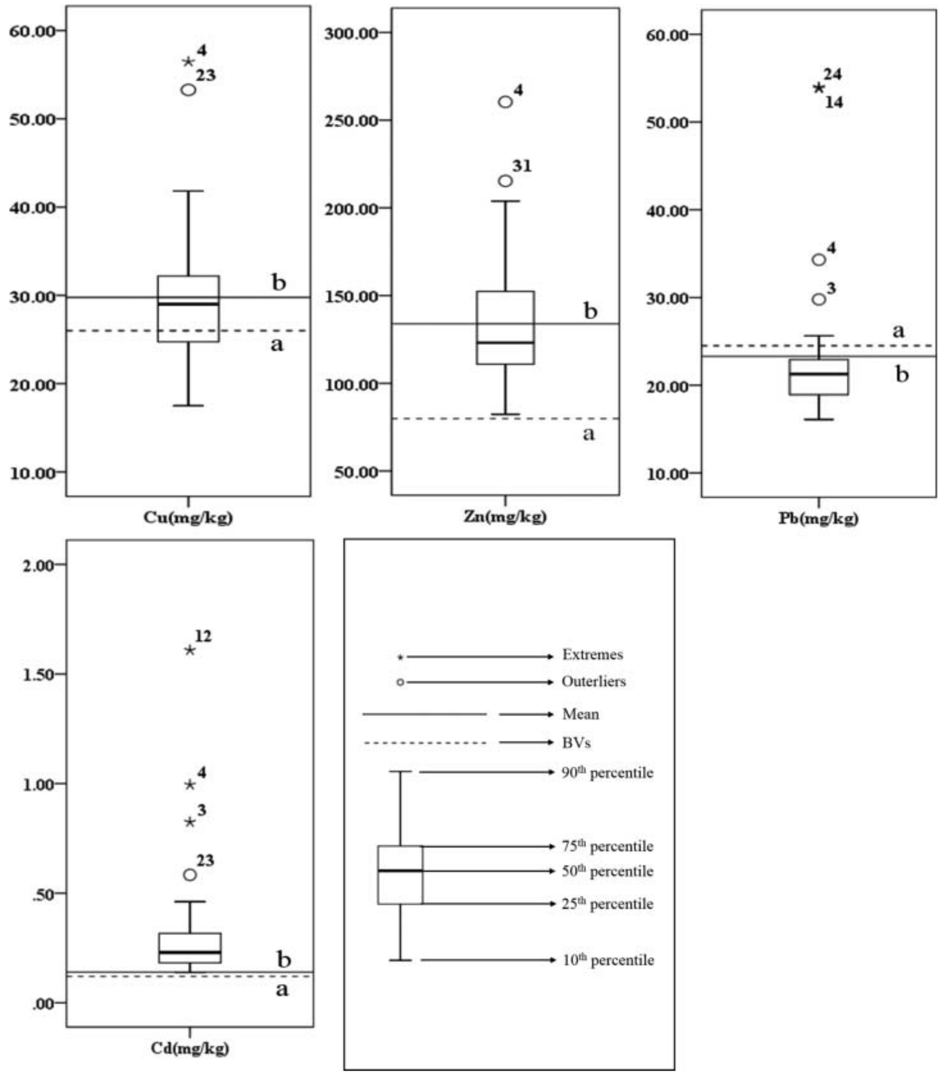

Figure 2. Boxplots of the HMs concentrations (mg/kg) in ORG soil (n = 34, BVs: the background values of Shanghai, the different letters "a" and "b" present significant difference, $p < 0.05$).

0.6 mg/kg for Cu, Zn, Pb, and Cd, respectively, the values of all HMs concentrations were significantly lower than GBs in this study. As shown in Figure 2, the mean concentrations for Cu, Zn, and Cd were significantly higher than their corresponding BVs, which indicated that soils in ORG may be contaminated by them. Though the mean Pb concentrations are significantly lower than their corresponding BVs, there were some sites' concentration were higher than BVs, and even had one extreme value, this may be due to its spatial heterogeneity. For all the HMs, the total concentrations showed different degrees of variability, and there were some outliers or extreme values for all the HMs, especially for Cd, reflecting the heterogeneous distribution of concentrations of anthropogenically emitted HMs (Liu *et al.* 2016).

Table 3. Mean concentration of HMs in different areas (mg/kg).

Sample location	Region/country	Cu	Zn	Pb	Cd	Reference
Greenbelt	Shanghai/China	30.02	133.37	51.05	0.32	Present study
Forest land/Chongming	Shanghai/China	17.40	66.20	13.40	0.13	Zheng *et al.* (2016)
Farmland soil	Shanghai/China	31.10	92.10	23.20	0.16	Long *et al.* (2013)
Greenbelt	Nanjing/China	38.80	113.40	53.10	0.11	Ding *et al.* (2011)
Roadside	Beijing/China	29.70	92.10	35.40	0.22	Chen *et al.* (2010)
Greenbelt in Traffic area	Harbin/China	29.05	110.59	39.27	0.24	Lu *et al.* (2012)
Forest shelter belt	Saransk/Russia	38.60	131.85	79.47	—	Kargin *et al.* (2014)
Green space	Auckland/New Zealand	20.50	56.50	42.50	0.17	Currancournane *et al.* (2015)
Forest areas and urban parks	Aliaga/Turkey	55.70	933.00	237.00	2.00	Odabasi *et al.* (2016)
Nationwide	Canadian	63.00	200.00	70.00	—	CCME[a] (1999)
Nationwide	AFNOR	100.00	300.00	100.00	2.00	AFNOR[a] (1996)
Nationwide	European	50–140	150–300	50–300	1.0–3.0	IEEP[b] (2009)

Note: Value of HMs in Auckland were mean of 0–10 cm and 10–20 cm. [a]Akoto *et al.* (2016); [b]Aydi (2015).

Comparison with urban forests/green lands and environment standards in different regions

The results presented in this study were compared with previous studies conducted in rural forests and farmland in Shanghai, noting that the concentrations of HMs in the ORG soil samples were higher than those in Chongming forest and farmland in Shanghai (except Cu) (Table 3) (Long *et al.* 2013; Zheng *et al.* 2016). The results may suggest that the ORG's soils were more affected by traffic. However, Chongming is an island, claimed to be the "last virgin territory" in Shanghai and located far away from urban areas, with relatively seldom influence by industrial activities (Deng *et al.* 2015). Additionally, the farmland is better protected due to its important function. Compared with urban forests/greening lands in other Chinese cities, the soil HM concentrations in ORG's soil samples were found to be more polluted than the soils of Nanjing, Beijing, and Harbin, with the exception of the level of Cu and Pb concentrations in the greenbelt of Nanjing (Chen *et al.* 2010; Ding *et al.* 2011; Lu *et al.* 2012). With regards to studies conducted in other countries, the soil HM concentrations in ORG were less concentrated than the results of Turkey and Russia's forests soils (except Zn), but higher than those of New Zealand (Currancournane *et al.* 2015). Compared with standard or guideline values of non-polluted soils in other countries, soil HM concentrations in ORG were less than the standard values proposed by the European norm for non-polluted soils, the French association of normalization, and Canadian environment quality guidelines (Akoto *et al.* 2016; Aydi 2015; Kargin *et al.* 2014; Odabasi *et al.* 2016).

Temporal variation of heavy metals in tree rings in the ORG

As shown in Figure 3, there is a general trend of increasing pollution with time. The mean concentrations of Cu, Zn, and Pb in the tree rings are the highest in 2013 (Cu. 75.62 mg/kg; Zn 26.43 mg/kg; Pb 1.21 mg/kg) compared with 2009–2012 (Cu, 50.11 mg/kg; Zn, 21.41 mg/kg; Pb 1.15 mg/kg) and 2004–2008 (Cu, 34.46 mg/kg; Zn, 14.64 mg/kg; Pb 1.14 mg/kg). Especially, the concentration of Cu in 2013 was 2.17 times higher than that in 2004–2008. The mean concentration of Cd (2013, 0.09 mg/kg; 2009–2012, 0.08 mg/kg; 2004–2008, 0.10 mg/kg) presented fluctuate change among different periods. The changes of HM concentrations did not differ significantly in the three different periods (except Cu) in spite of the observed trend, and this result was similar to the result of Cocozza *et al.* (2016). Heavy metals in plant samples have also been

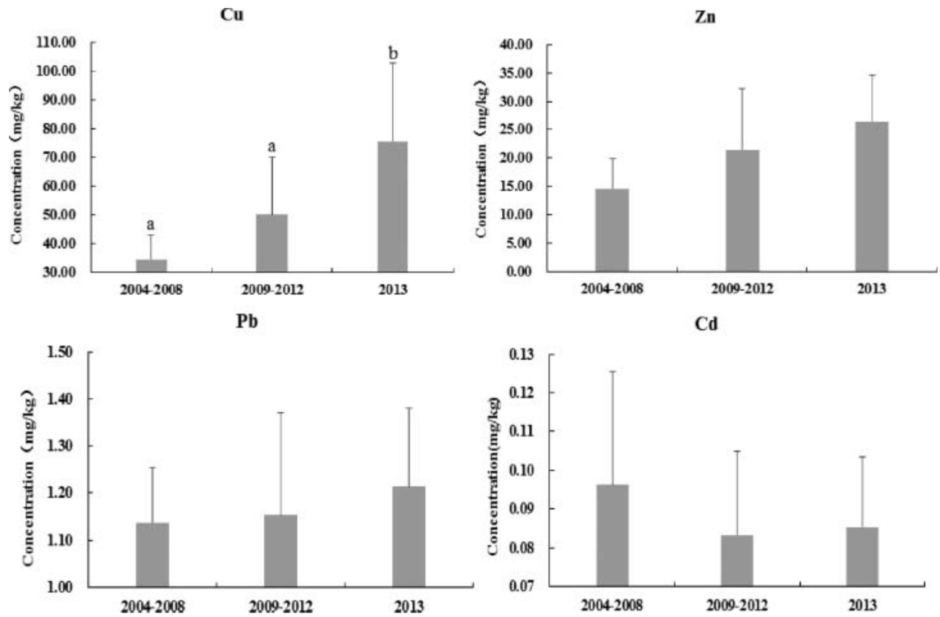

Figure 3. Concentrations of HMs in tree-ring (mg/kg n = 7 mean values ± standard deviation). Note: Values with different letters "a" and "b" present significant differences between different periods (one-way ANOVA, $p < 0.05$).

measured and reported around the world. Compared with previously published reports, concentrations of Cu, Zn, Pb, and Cd (53.4, 20.83, 1.17, and 0.09 mg/kg) were higher for this study than those in tree rings from Venafro Plain in Italy (Zn, about 2.5 mg/kg; Cu, Pb, and Cd were non-detected) (Cocozza *et al.* 2016), and in plant stems in Aliaga industrial region in Turkey (Cu,0.93 mg/kg; Zn, 11.9 mg/kg; Pb, 0.1 5 mg/kg; Cd, 0.13 mg/kg) (Odabasi *et al.* 2016). Different results may be due to different parts of plants, different species, and different environments tested. In addition, according to a previous study, the normal concentrations of Cu, Zn, Pb, and Cd in plants are 3–10 mg/kg, 10–150 mg/kg, 0.5–10 mg/kg, and 0.05–2 mg/kg (Padmavathiamma and Li 2007); values below or above this range indicate deficiency or phytotoxicity for plants. Literature data indicate that as Pb concentration in soil increases, the content of Pb in plants changes accordingly, and Cu behaves the same way (Sieciechowicz *et al.* 2014). However, it may be different for different species or different elements. The results obtained in our study indicated that *C. camphora (L.)* contained the highest concentrations of Cu, was 5.34 times of the upper limit of normal concentration of Cu in plants (10 mg/kg) as suggested by Padmavathiamma and Li (2007), which could possibly be used as an accumulation plant in soil phytoremediation for Cu. Different species have different tolerances for HMs, depending on the plant species, as well as on the growth stage (Alahabadi *et al.* 2017); for better use of *C. camphora (L.)* in phytoremediation, we should conduct further studies in the future.

Pollution assessment

I_{geo} of heavy metals

As shown in Figure 4, compared with BVs, the mean I_{geo} of Cu (−0.44) and Pb (−0.72) were lower than 0, which indicated the soil were unpolluted by Cu and Pb. The mean of Zn (0.11) and

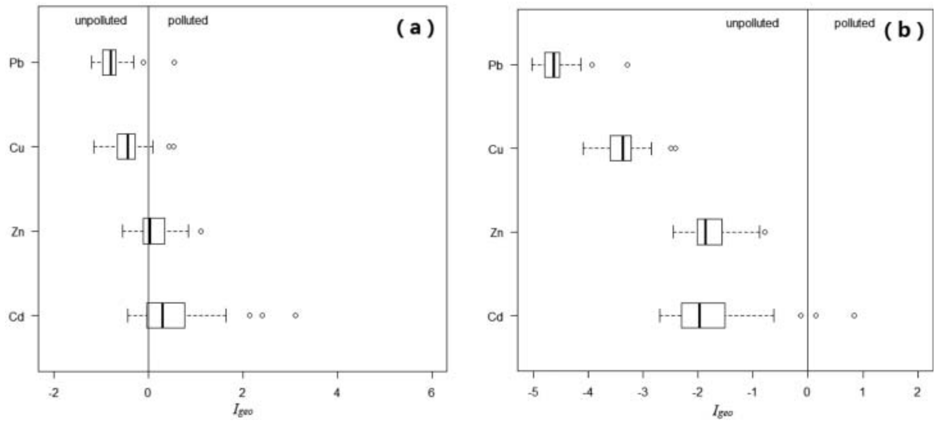

Figure 4. Assessment for HM in ORG using I_{geo} (n = 34). (a) Present the reference values was BVs and (b) present the reference values was GBs. The mean of every element in this figure is same to Figure 2.

Cd (0.51) were higher than 0, which suggested the soil pollution level of Zn and Cd were unpolluted to moderately polluted. When compared with GBs, the values of I_{geo} were less than 0 for all HMs, except for Cd in two samples, which suggested that the soils in ORG were slightly polluted with HMs (Cu, Zn, Pb, and Cd) according to the soil quality in China (Grade II).

Potential ecological risk index of heavy metals

According to the BVs, the results of mean potential ecological risk of single HM (*EI*) were ranked as Cd (78.60)>Cu (5.73)> Pb (4.76)>Zn (1.62), indicating that Cd presented a moderate risk in the soil, whereas other HMs (Cu, Zn, Pb) showed low risk (Table 4), but the mean ecological risk index of all HMs (*RI*) indicated that all sites in ORG were at low risk level. Compared with GBs as references, the mean ecological risk of single HMs was ranked: Cd (16.26)>Cu (0.74)>Zn (0.45)>Pb (0.33) (Table 4), which indicates low ecological risk for each HM. For the potential ecological risk of all HMs (*RI*), all the sampled sites have low potential ecological risk.

Source identification of the heavy metals in soil and tree-ring

Relationship between heavy metals and soil properties

In order to understand the interrelationships between different HM, and the effect of soil properties on the availability of soil HMs, HM-HM and HM-soil properties Pearson correlations were

Table 4. Potential ecological risk indices in different sites (n = 34).

	EI					*EI*				
Item	Cu[a]	Zn[a]	Pb[a]	Cd[a]	*RI*[a]	Cu[b]	Zn[b]	Pb[b]	Cd[b]	*RI*[b]
Mean	5.73	1.62	4.76	78.60	90.71	0.74	0.45	0.33	16.26	17.78
Min	3.37	0.82	3.29	33.40	43.58	0.44	0.27	0.23	6.91	8.23
Max	10.86	3.26	11.02	388.86	399.30	1.41	0.87	0.77	80.43	81.78
SD	1.59	0.45	1.76	70.80	71.73	0.21	0.13	0.12	14.64	14.79
CV (%)	27.71	28.00	36.93	90.08	79.08	27.71	29.28	36.93	90.08	83.19

Note: "a" presents the reference values was BVs, and "b" presents the reference values was GBs.

Table 5. Correlation coefficient of soils HM concentrations and soil properties (n = 34).

Item	pH	TN	TP	OM	Cu	Zn	Pb
Cu	−0.22	0.427*	−0.21	0.05	1.00		
Zn	−0.390*	0.31	−0.09	−0.02	0.616**	1.00	
Pb	−0.17	0.33	−0.01	−0.08	0.448**	0.18	1.00
Cd	−0.410*	0.12	0.04	−0.15	0.349*	0.604**	0.351*

Note: *Correlation is significant at $p < 0.05$, **Correlation is significant at $p < 0.01$, Dark gray: r > |0.5|, light gray: |0.25| < r < |0.5|, and the same meaning in Table 6.

calculated (Table 5). A significant negative correlation has been observed between Zn and Cd with soil pH. Previous studies showed that pH presented a significant positive correlation with concentration of Cd (Alamgir s*et al.* 2015; Nan *et al.* 2002), while another study showed Cu, Zn, and Cd were negatively correlated with soil pH (Kirkham 2006; Zhao *et al.* 2010), and Jadzevi et al.'s (2014) study showed a very weak correlation between the soil pH and concentration of HMs. The different results may demonstrate that soil HMs present different correlations with pH under different conditions, indicating that the correlation between soil HMs and pH is complex. In this study, concentration of Cu showed significant positive correlation with soil TN; this may indicate that TN plays an important role in Cu accumulation. Cu is most likely to originate from anthropogenic sources like soil TN. Contrary to the results in previous studies, which suggest that soil OM is one of the most important soil properties affecting heavy metal availability (Antoniadis *et al.* 2008; Zhao *et al.* 2010), there were no significant correlations between OM with soil HMs in this study. Moreover, soil TP showed no significant correlation with HMs. Above all, the correlation between soil properties and HMs present great complexity, and further research is needed.

Apart from soil properties, the HMs themselves are correlated with one another (Table 5). The results showed that Zn, Pb, and Cd has significant positive correlation with Cu, therein, Zn and Cu performed extreme positive correlation. In addition, Zn and Cd showed extreme significant positive correlations. According to the previous researches, HMs that have high positive correlations may reflect that these heavy metals had common source (Cai *et al.* 2012; Franco Uría *et al.* 2009).

The influence of soil properties and soil HMs concentrations on HMs concentrations in tree rings

Correlation analyses were performed to identify the influence of soil properties and HM concentrations in soil on HM concentrations in tree rings. Soil pH was significantly and negatively correlated with the concentration of Cd in tree rings, and concentration of Cu, Zn, and Cd in soil was significantly and positively correlated with the concentration of Cd in tree rings (Table 6). While no significant correlations were found between Cu, Zn, and Pb in

Table 6. Correlation coefficients of HM concentrations in tree rings and soil and soil properties (n = 7).

Item	pH^S	TN^S	TP^S	OM^S	Cu^S	Zn^S	Pb^S	Cd^S
Cu^T	0.054	−0.638	−0.467	−0.147	0.284	0.355	0.317	0.426
Zn^T	0.453	−0.067	−0.602	−0.325	−0.241	−0.456	−0.070	−0.447
Pb^T	−0.307	0.570	0.124	0.337	−0.024	0.041	−0.203	0.061
Cd^T	−0.805*	0.075	0.505	−0.033	0.838*	0.818*	0.713	0.862*

Note: T present in tree rings, S present in soil.

tree rings with soil properties and HM in soil. These results suggested that lower pH will promote the accumulation of Cd in tree rings from soil, and this finding was in agreement with previous reports (Zeng *et al.* 2011). On the other hand, the concentrations of Cu, Zn, and Pb in tree rings were not influenced by soil properties and HM concentrations, raising the possibility that these elements are not necessarily incorporated into the tree tissue from the soil. Perhaps they are absorbed from the air (Kirchner *et al.* 2017). Further research aimed at identifying Cu, Zn, and Pb concentrations should be conducted, in order to distinguish between soil uptake and aerial interception.

Literature data indicate that as Pb concentrations in soil increase, the content of Pb in plants changes accordingly, and Cu behaves the same way (Sieciechowicz *et al.* 2014). In this study, there were significant positive correlations between concentration of Cd in tree rings with concentration of Cu, Zn, and Cd in soil, which indicated that Cd in tree rings mainly uptake from Cd in soil, and the Cd in tree rings may have common source with Cu and Zn in soil. Moreover, there were no significant correlations between Cu, Zn, and Pb in tree rings with soil properties and HMs in soil, and this may suggest that the HMs concentrations in tree rings are assimilated in another way (Panyushkina *et al.* 2016). As previous studies reported, plants have the ability to uptake HMs from soils, water, and air through roots, barks, and leaves (Cocozza *et al.* 2016; Mahar *et al.* 2016). Although tree-ring analysis has the ability to reveal some of the cause–effect relationships, it is important to remember that tree-ring is the sum of a whole suite of processes acting on a tree. HMs in tree rings may be absorbed from acid rain, surface water uptake, and air pollution through roots, barks, and leaves, and HMs concentrations detected in tree rings being dependent on the release mode of contaminant, contaminant concentrations, and tree species (Balouet *et al.* 2007; Innes and Cook 2011). As the analysis results showed in the identification of sources of HMs in soil, Cu, Zn, and Pb in tree rings are much more likely come from traffic, mainly vehicle exhausts or abrasion of vehicle parts of tires. In brief, the current results indicate that the sources of HMs in tree rings are variable. Therefore, plant physiological functions should be taken into consideration for precisely predicting heavy metal concentrations in tree rings.

Principal component analysis

The results of PCA by applying Varimax rotation with Kaiser Normalization for HMs concentrations and soil properties are shown in Table 7. Three principal components (PC) with eigenvalues higher than 1 (before and after rotation) were extracted. The graphic representation of the three components is also shown in Figure 5, where the associations between HMs and soil properties can be seen. The results indicate that PCA leads to a reduction of the initial dimension of the data set to three components, which explain a 64.9% of the data variation. The first principal component (PC1) explains 26.3% of the total variance, HMs of Cd, Zn, and Cu were grouped into the soil properties of pH. The Cd, Zn, and Cu loadings were 0.84, 0.83 and 0.46, respectively, while pH loading was −0.65. In fact, the concentrations of Cd and Zn were significantly correlated with soil pH (Table 5). The second principal component (PC2), HMs of Pb and Cu were grouped into the soil properties of TN, which accounted for 21.4% of the total variance, while the concentration of Cu were significantly correlated with Pb (Table 5), and their loadings were Pb (0.75), Cu (0.65) and TN (0.79). However, third principal component (PC3) is correlated mainly with TP and OM, which has a loading value 0.80 and −0.78, accounting for17.2% of the total variance, implied that the soil properties of TP and OM did not correlate with any HMs (Cai *et al.* 2012).

Table 7. Total variance explained and matrix of principal components analysis (significant loading factors are remarked in bold) n = 34.

	Initial eigenvalues			Extraction sums of squared loadings			Rotation sums of squared loadings		
Component	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %
Total variance explained									
1	2.813	35.158	35.158	2.813	35.158	35.158	2.103	26.291	26.291
2	1.373	17.166	52.324	1.373	17.166	52.324	1.713	21.408	47.698
3	1.004	12.555	64.879	1.004	12.555	64.879	1.374	17.18	64.879
4	0.881	11.016	75.894						
5	0.746	9.326	85.22						
6	0.658	8.22	93.44						
7	0.315	3.938	97.378						
8	0.21	2.622	100						

	Component matrix			Rotated component matrix		
Items	PC 1	PC 2	PC 3	PC 1	PC 2	PC 3
Component matrix						
pH	**−0.614**	−0.002	0.271	**−0.646**	−0.173	0.059
TN	**0.594**	−0.043	**0.536**	0.126	**0.791**	−0.025
TP	−0.139	**0.778**	0.216	−0.160	0.050	**0.802**
OM	−0.053	**−0.792**	0.058	−0.160	0.044	**−0.778**
Cu	**0.775**	−0.213	0.200	**0.457**	**0.650**	−0.236
Zn	**0.795**	−0.011	−0.339	**0.827**	0.234	−0.089
Pb	**0.575**	0.116	0.510	0.143	**0.753**	0.132
Cd	**0.705**	0.281	−0.423	**0.84**	0.100	0.198

Note: Extraction method: principal component analysis; rotation nethod: varimax with Kaiser normalization.

Source of HMs in soil

The major source of high HM concentrations in the soils can be ascribed to metal rich source rocks, atmospheric pollution from motor vehicles, combustion of fossil fuels, agricultural fertilizers and pesticides, organic manures, disposal of urban, and industrial wastes, as

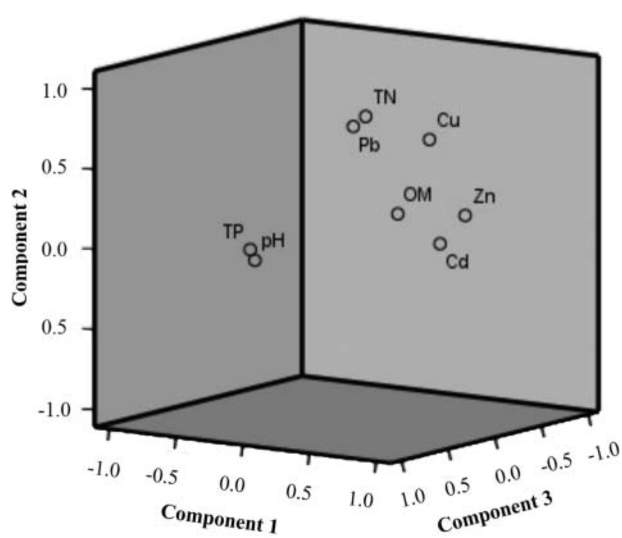

Figure 5. Loading plot of PC1, PC2 and PC3 (n = 34).

well as mining and smelting processes (Turer 2005). In this study, descriptive statistics showed that Cu, Zn, and Cd had higher concentrations compared to local background values (Figure 2), whereas Pb had relatively lower concentrations than its background values. This indicated that Cu, Zn, and Cd are more likely to have originated from an anthropogenic source, while the Pb may result from a natural source. The descriptive statistics (Figure 2), correlation analysis (Table 5), and PCA results (Table 7 and Figure 5) indicated that HMs in ORG may be categorized into two groups: (1) Cd and Zn; (2) Pb and Cu. In consideration of the complex relationships between soil HMs and environmental factors, we doubt whether this classification is correct, but, there is no doubt that the soil HMs concentrations were influenced by anthropogenic activities.

Many previous studies reported high concentrations of HMs especially in soils along highways (Turer 2005). Highways, as important elements of urban transportation, bear significant traffic pressure. The rapid urbanization process has brought a sharp growth of motor vehicles to Shanghai. In the past two decades, the civil motor vehicles in Shanghai increased from 370333 in 1994 (SBS 1995) to 3323500 in 2015 (SBS 2016). It has been previously reported that pollution by Cu, Zn, Pb, and Cd can be linked with vehicle traffic (Cambi *et al.* 2015; Duong and Lee 2011). Cu is often used in brake pads and radiators and is also added to automotive lubricants, ZnO and ZnS are added to the tire during vulcanization, tetraethyl lead is added in gasoline as antiknock agent, and braking systems could produce significant amounts of Cu, Zn, Pb, and a small amount of Cd (Adamiec *et al.* 2016; Chen *et al.* 2016). Although the use of unleaded petrol had been enforced, the content of Pb accumulated in soils can still persist for a long time (Chen *et al.* 2016). Moreover, it should be noted that higher vehicle speed and asphalt surface pavement could result in greater tire wear and increased fuel combustion, which can lead to more emission of HMs (Duong and Lee 2011; Murphy *et al.* 2015). The outer-ring highway, as an important part of Shanghai's traffic system, was constructed from asphalt and a mixture of asphalt and concrete and was designed for traffic speed of 80 km/h (Shi 2006; Xu 2004). Moreover, it is generally assumed that contaminations by traffic are of non-point source origin, coming mostly either from vehicle exhausts or from the abrasion of vehicle parts of tires.

Historically, agriculture was the first major anthropogenic activity on the soil (Chen *et al.* 2016). As a metropolis with limited land resources, the increase of forest in Shanghai was most converted from farmland. Similar with many cities, fertilizers, waste water and solid waste have been widely used by industrial enterprises to improve farmland soil fertility in Shanghai before 1992. Cd and Zn are mainly related to their use in the agricultural soils of Shanghai (Long *et al.* 2013). In fact, long-term and intensive use of fertilizers, waste water, and solid waste may lead to serious HMs contamination in soils even after discontinuing for approximately two decades (Sieciechowicz *et al.* 2014; Zhao *et al.* 2009). In addition, the use of various agrochemicals and pesticides usually contain Cd, and they may also be an important source of Cd to soil (Cai *et al.* 2012; Long *et al.* 2013). TN seems to play an important role in Cu accumulation (Tables 6 and 7), indicating a common origin of Cu and nitrogen due to the application of different fertilizers.

Moreover, rapid population growth, urbanization, industrial growth, and economic development could lead to the generation of significant quantities of solid wastes that are causing serious environmental degradation (Kamal *et al.* 2016). Though the use of solid waste to improve farmland soil has been forbidden, as a fast developing megacity, the solid waste generation rate is increasing quickly. In order to solve this problem, some waste solids were ending in landfills. Furthermore, as one of the large scale ecological programs in Shanghai, ORG had large demands for soil, therefore, part of the soil in ORG was formed from solid waste landfill. However, untreated solid wastes usually contain high concentrations of HMs and can seriously pollute the soil even many years later (Aydi 2015).

Conclusions

1) Concentrations of Zn and Cd in soil were significantly higher than the BVs. Although soil concentrations of Cu was higher than BVs, there were no significant difference between concentrations of Cu and Pb. All the HMs concentrations surveyed in this study were significantly lower than the GBs. Concentrations of Cu, Zn, and Pb in tree rings increased gradually in the past 10 years, and the concentration of Cu increased significantly in 2013, but the concentrations of Cd follow an irregular change.
2) Integrating the results of I_{geo} and the Potential Ecological Risk Index: compared with BVs, showed that Cu and Pb were in an unpolluted level, Zn and Cd were in unpolluted to moderately polluted level, the *EI* were all in low level except for Cd, which was classified to a moderate level. Cd should be identified as a control priority, but the RI of all surveyed sites were in low levels, compared with GBs, all the HMs were in unpolluted level and have low ecological potential risk.
3) Concentrations of HMs were influenced by the extrinsic factors. Correlation analysis and PCA results indicated that the origin of HMs in soil and tree rings were associated in a complex manner, the concentrations of Zn and Cd decrease as soil pH increase, concentrations of soil TN promoted the accumulation of Cu; vehicle exhausts and abrasion of vehicle parts of tires produced in traffic maybe the main source of Cu, Zn, and Pb, and fertilizers, waste water, and solid waste used in agriculture soil maybe the sources of HMs, and Cd were mainly originated from fertilizer and pesticides include Cd; Cd in tree rings were mainly uptaken from the soil, while the other HMs may be uptaken from surface water uptake and air pollution.
4) *C. camphora (L.)*., as a bioindicator, has the potential to reconstruct the change of Cu, Zn, and Pb, but the correlation between concentrations of HMs in the tree rings and soil need further study, and the source of HMs in its tree-ring needs to be identified further.

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Conflicts of interest

The authors declare no conflict of interest.

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