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Intra-urban biomonitoring: Source apportionment using tree barks to identify air pollution sources

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ABSTRACT

It is of great interest to evaluate if there is a relationship between possible sources and trace elements using biomonitoring techniques. In this study, tree bark samples of 171 trees were collected using a biomonitoring technique in the inner city of São Paulo. The trace elements (Al, Ba, Ca, Cl, Cu, Fe, K, Mg, Mn, Na, P, Rb, S, Sr and Zn) were determined by the energy dispersive X-ray fluorescence (EDXRF) spectrometry. The Principal Component Analysis (PCA) was applied to identify the plausible sources associated with tree bark measurements. The greatest source was vehicle-induced non-tailpipe emissions derived mainly from brakes and tires wear-out and road dust resuspension (characterized with Al, Ba, Cu, Fe, Mn and Zn), which was explained by 27.1% of the variance, followed by cement (14.8%), sea salt (11.6%) and biomass burning (10%), and fossil fuel combustion (9.8%). We also verified that the elements related to vehicular emission showed different concentrations at different sites of the same street, which might be helpful for a new street classification according to the emission source. The spatial distribution maps of element concentrations were obtained to evaluate the different levels of pollution in streets and avenues. Results indicated that biomonitoring techniques using tree bark can be applied to evaluate dispersion of air pollution and provide reliable data for the further epidemiological studies.

1. Introduction

Air pollution has a severe adverse effect on human health. Only 12% of world population resides in cities with air pollution levels below the established by World Health Organization (WHO) guidelines (WHO, 2014). Air pollution can originate from numerous sources, such as power plants, steel plants, wood burning, sea salt, cooking exhausts, vehicular traffic, dust, vegetal parts and decomposition and manufacturing facilities (Hu et al., 2014; Masri et al., 2015). However, the main source of air pollution in large cities is vehicular emissions including tailpipe and non-tailpipe emissions. Vehicular emissions are associated with light or heavy vehicle fleets, fuel, age of the vehicle, maintenance, condition of the vehicle, speed and other variables (Toledo and Nardocci, 2013). The extent of vehicular emissions depends also on traffic levels, infrastructure design, driving patterns and vehicular characteristics (Sharma et al., 2010). The elements emitted by traffic can be different with various processes, such as incomplete fuel burning, tire and brake wear-out processes, and road dust resuspension (Schauer et al., 2006). Abu-Allaban et al. (2003) found that major emission of particulate matter (PM) smaller than 10 μm (PM_{10}) came from brake wear-out and road dust resuspension, while PM smaller than 2.5 μm ($PM_{2.5}$) came from vehicular tailpipe emissions. The PM (mostly $PM_{2.5}$ and PM_{10}) derived from vehicular emissions can penetrate deeply into the lungs and heart causing damage (Nel, 2005).

Trace elements, mainly heavy metals can affect human health including genotoxic effects, cancer induction, and damage to the immune and neurological systems (Schauer et al., 2006). Air pollution is described as one of the causes of decrease in life expectation (Pope et al., 2009), causing the high risk of cardiovascular and lung diseases (Langrish and Mills, 2014; Raaschou-Nielsen et al., 2011). According to Mills et al. (2009) acute exposure can cause a range of adverse cardiovascular events, as evidenced by hospital admissions due to angina, myocardial infarction, and heart failure. Epidemiological studies from Sao Paulo City have evaluated the effects of air pollution on human health using data measured at monitoring stations. However, these monitoring stations are not able to cover the total study area and do not measure metal elements either. Furthermore, air pollution presents a heterogeneous dispersion in urban areas but it is usually assumed that the pollution is homogeneous in epidemiological studies (Schauer et al., 2006).

Biomonitoring can be a technique that provides the information on elemental composition and spatial characterization with relatively low

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cost in urban atmospheres. A biomonitoring study using *Tillandsia usneoides* in Sao Paulo City found the highest concentrations of Zn and Ba on the avenue with heavy duty traffic (Figueiredo et al., 2007). Carneiro et al. (2011) observed that Al, S, Cl, V, Fe, Cu, and Zn showed higher concentrations close to traffic corridors. The measurements of tree barks, as a biomonitoring technique, is already well established in order to estimate air pollution levels in urban areas (Guéguen et al., 2012; Perelman et al., 2010; Catinon et al., 2009). This study demonstrates the feasibility of using tree barks for source apportionment and construction of intra-urban microscale maps.

The objective of this study was to determine trace element in tree barks collected at different street classifications in urban areas, and to identify the plausible sources of pollution in the inner city of Sao Paulo. Our hypothesis was if tree bark can be used to identify the emission of chronic local atmospheric pollution (<500 m from streets) and if the different spatial distributions of vehicle-induced elements are correlated with vehicular emissions.

2. Methods

2.1. Study area

Samples of tree barks were collected at inner city area of São Paulo (Fig. 1), which is located at 23°32′51″ S and 46°38′10″ W and has a total area of approximately 150 km^2. A total of 171 tree bark samples (4 species) were collected in 75 different streets and avenues. A total number of 22 samples of tree barks were collected at the control site. The control site is a site without vehicular traffic inside a forest reservation located at 23°31′49″ S and 47°06′14″ W and 64 km from São Paulo City.

According to the Company of Traffic Engineering (CET) of São Paulo City, this city has approximately seven million registered vehicles. On average, traffic flow is composed of 79% of light vehicles, 16% of motorcycles, 2% of heavy vehicles and 4% of buses (CET, 2013). The city has the driving restriction based on a vehicle's license plate number since 1997. The vehicles restricted are not allowed to enter the inner city between 7 am and 10 am, and between 5 pm and 8 pm. The official classification of the streets in the Sao Paulo City by CET is: Express, Arterial, Collector, and Local, based on traffic density (CET, 2013).

2.2. Tree bark sampling and preparation

Tree barks were randomly sampled at main roads, streets, and local streets with a relatively low traffic density in the inner city. The tree species collected were *Tipuana tipu* (40.80%), *Caesalpinia pluviosa* (38.46%), *Ligustrum* sp. (13.02%), and *Tibouchina granulosa* (7.69%), which are the most common species at the streets of São Paulo. We avoided collecting bark samples from trees that were < 10 m apart. It was prioritized to collect in a dry season in order to avoid degradation of the collected samples. Each sample was placed into a paper bag and stored at a cabinet under a low humidity until analysis. Information on tree species, location, geographical coordinates, and land use was recorded.

At the laboratory, the collected tree barks were first cleaned using a soft nylon dental brush to eliminate mainly external materials, such as dead insects, dust and lichens. The bark samples were obtained by grating the external layers of 3 mm thickness and used all this material after a homogenization. A titanium grater and a caliper ruler to measure the 3 mm thickness were used to obtain the material. Each sample was then ground to a powder using a mill with agate mortar. The powder was compressed by applying a pressure of 15 tons to make a double layer pellet composed of 0.5 g of tree bark powder and 1 g of boric acid (H_3BO_3, puriss p.a. ACS reagent).

2.3. Elemental analysis

The bark samples prepared in pellets were analyzed using EDXRF (EDX 700-HS, Shimadzu Corp., Kyoto, Japan). The concentrations of elements (Al, Ba, Ca, Cl, Cu, Fe, K, Mg, Mn, Na, P, Rb, S, Sr and Zn) were calculated by the method of fundamental parameters, converting the intensity to quantitative values of the elements. The certified reference material, National Institution of Standards and Technology (NIST) #1547 Peach Leaves, were also analyzed using the same procedure of tree bark samples to evaluate the accuracy and precision.

2.4. Data analysis

Kolmogorov–Smirnov normality test ($P < 0.0001$) was applied to determine if the element concentrations followed a normal distributions. Kruskal–Wallis test ($p < 0.05$) was also used to compare the means element concentrations obtained for each type of street classification. The discriminant analysis, a multivariate statistical method used to discover the characteristics that distinguish a group of members to another one, was used to classify sampling sites according to emission sources. The street classification presented by CET (2013) was compared with one obtained by applying discriminant analyses in our results, based on how well each classification predicted the test results.

Principal Components Analysis (PCA) using Varimax rotation method with Kaiser normalization was applied to obtain the correlations between the element concentrations and to determine their possible origins. The statistical analyses were conducted using SPSS statistical package version 17 and established for 95% confidence intervals.

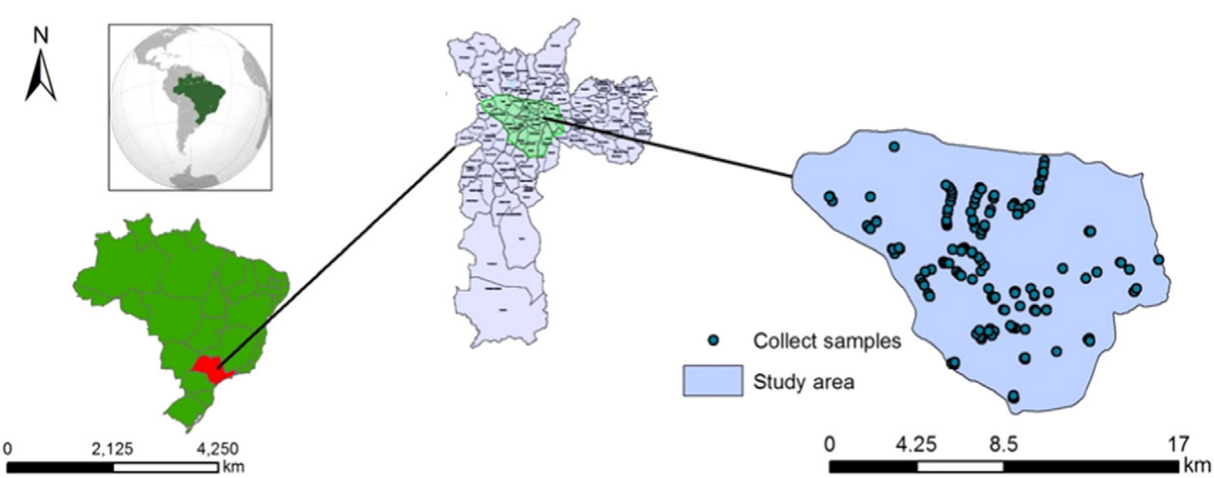

Fig. 1. Map of the study area in São Paulo City; the points indicate the locations of collected tree bark samples.

Table 1
Elemental concentration statistics (mg/kg).

Element	Number of samples	Median	Minimum value	1 quartile	3 quartile	Maximum value
Al	193	579	57	315	1019	6819
Ba	193	444	22	207	845	21,089
Ca	193	32,900	1070	23,571	39,391	59,628
Cl	192	78	7	47	145	21769
Cu	193	6	4	5	8	28
Fe	193	778	61	427	1772	7356
K	193	1163	278	937	1673	12,473
Mg	193	1161	110	596	2049	4055
Mn	193	39	10	28	62	183
Na	193	20	4	17	23	127
P	193	938	90	802	1075	2078
Rb	193	12	4	10	15	67
S	193	2880	1243	2318	3634	9148
Sr	193	102	14	63	142	316
Zn	193	96	7	39	184	599

The Ordinary Kriging method was used to construct microscale element concentration maps. These Microscale maps were obtained for elements associated with vehicular emissions according to the PCA results. The error maps were created for the validation of the test. The Kriging analyses were conducted using ArcGIS software version 10.0.

3. Results

The statistics of elemental concentrations are presented in Table 1. The highest concentration of the elements was Ca, followed by S, K, Mg and Fe. The results of the Kolmogorov–Smirnov normality test indicated that the data do not follow a normal distribution ($P < 0.0001$). The Kruskal–Wallis test ($p < 0.05$) showed significant differences in the mean value of elements between each street classification except for Rb and Na. The hierarchy of sum of element concentrations and the pollution radar chart of each element concentration by the CET street classification are shown in Fig. 2-a and -b, respectively. Table 2 represents the difference in the number of streets classified by the CET street classification and new proposed classification from the discriminant test.

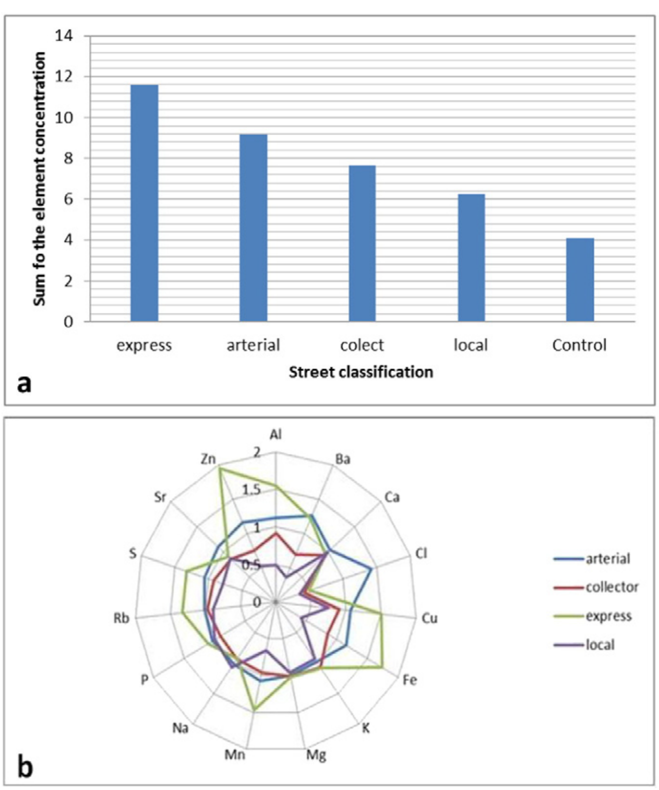

Fig. 2. Elemental concentrations (mg/kg) by the CET street classification as a function of traffic density: a) sum of mean elements concentrations; and b) pollution radar chart of each element concentration.

Table 2
Difference in the number of streets classified by the CET street classification and new proposed classification.

			Predict street classification				
			Arterial	Collect	Express	Local	Total
CET street classification	N	Arterial	**35**	21	19	24	99
		Collect	11	**15**	3	13	42
		Express	4	1	**1**	1	7
		Local	2	8	0	**13**	23
	%	Arterial	**35.4**	21.2	19.2	24.2	100
		Collect	26.2	**35.7**	7.1	31	100
		Express	57.1	14.3	**14.3**	14.3	100
		Local	8.7	34.8	0	**56.5**	100

According to PCA statistical results, 73.6% of variance could be explained by five components (Table 3). The 1st component is comprised of Al, Ba, Fe, Mn, Cu, and Zn; the 2nd includes Ca and Sr; the 3rd includes Cl and Na; the 4th includes K and P, and the 5th is characterized by Mg and S. The predictive maps of selected elements are presented in Fig. 3.

4. Discussion

The hypothesis of this study is that the determination of the element concentrations in tree bark samples can be a useful tool for accurate estimates of particulate pollutants from vehicle emissions. Our results demonstrated that there is a hierarchy of element concentrations according to the street classification by traffic density. The streets classified as express presented the highest sum of elemental concentrations and the streets classified as local showed lowest sum of elemental concentrations, as shown in Fig. 2. In addition, our results demonstrate that chemical analyses of tree barks enable the identification of different hotspots in the concentrations of traffic related elements in a given street classification, based on the locations of sample collection.

From Table 2 the discriminant analysis indicates that some points of the streets cannot be adequately classified with the official classification determined by traffic density. A 63% of studied points had different classifications, based on emissions, from the official classification. The 99 points classified as arterial streets became 35 arterial streets, 21 arterial collect, 19 arterial express and 24 arterial local. Note however, that

Table 3
Principal Components Analysis using Varimax rotation method with Kaiser normalization.

	Components				
	1	2	3	4	5
Al	**0.853**	−0.175	−0.098	0.048	−0.078
Ba	**0.548**	0.004	−0.015	0.109	−0.333
Ca	−0.239	**0.915**	−0.002	0.027	−0.144
Cl	0.034	0.008	**0.933**	−0.033	0.082
Cu	**0.741**	0.138	−0.001	−0.068	0.247
Fe	**0.918**	−0.125	−0.109	0.067	0.063
K	0.054	−0.434	0.009	**0.721**	0.303
Mg	0.105	−0.067	−0.027	0.189	**0.847**
Mn	**0.711**	−0.145	−0.097	0.253	0.104
Na	−0.202	−0.019	**0.912**	−0.026	−0.094
P	0.041	0.355	−0.106	**0.813**	−0.124
Rb	0.41	−0.128	0.044	0.43	0.175
S	0.493	0.389	0.057	−0.136	**0.532**
Sr	0.086	**0.889**	−0.007	−0.013	0.149
Zn	**0.779**	0.172	0.013	0.002	0.289
Eigenvalue	4455	2253	1744	1443	1138
% of variance	27,107	14,871	11,676	10,072	9831
Cumulative %	27,107	41,978	53,654	63,726	73,557

Bold data represent the element that are component of the source in the row.

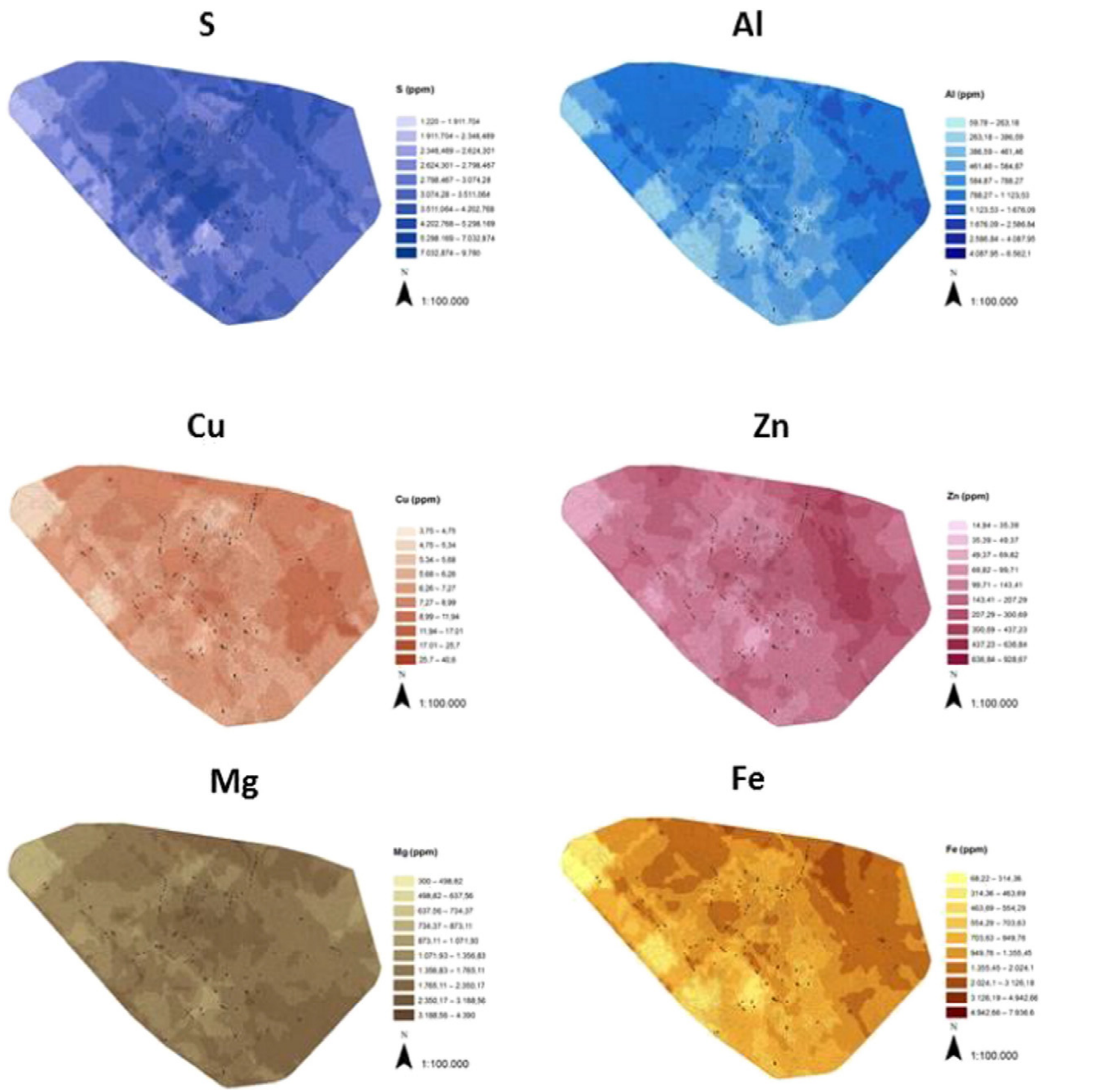

Fig. 3. Predictive spatial maps of the element resuspension dust and vehicular emission (Al, Cu, Fe, and Zn) and fossil fuel combustion (Mg and S) by the PCA results.

other factors such as squares, vertical obstacles, median strip with trees, garages, and parking spaces are more likely affect the local characterization and the local exposure profile. In our study, some samples from trees located near garages at the street with low volume traffic showed similarity to the emission characteristics at the streets with high volume traffic. On the other hand, some samples collected from trees located at squares and at median strip with high volume traffic showed similarity to the emission characteristic at the streets with median or low volume traffic. In these cases, the elemental profile related with traffic was different from the street classification by traffic density. This discrepancy may indicate an important role in establishment of the relationship between urban air pollution and human health because adequate exposure assessment is essential for accurate human health risk assessment.

Our results also demonstrate that the methodology using tree barks as a biomonitor can be applied to identify the specific emissions sources, in particular, as a marker of vehicle emissions in this study. The higher concentrations of elements found in tree barks were Ca, S, Mg and Fe, which are also found with higher concentrations in earth crustal and road dust, indicating the impact of road dust resuspension in the study area (Sternbeckc et al., 2002). Using PCA statistical analysis, the possible sources are identified. Group 1 is characterized by vehicle emissions mainly on express roads, which explains 27.1% of the total variance. Elements included in this group are primarily related to resuspension of road dust (Al, Fe and Cu) and wear-out of brakes and tires (Ba, Cu and Zn). Several studies have already shown that the vehicle emissions are a main source of air pollution in the Sao Paulo City (Martins et al., 2006; Sánchez-Ccoyllo et al., 2008; Andrade et al., 2012). Group 2 is characterized with Ca and Sr, which explains 14.8% of the total variance. Yeung et al. (2003) suggested that the source of Ca and Sr could be pavements materials in Hong Kong City. Group 3 (Cl and Na) explains 11.6% of the variance. Although Sao Paulo City is approximately 60 km distant from the sea coast, sea salt can affect the city, depending on the prevailing wind direction. During this study, the prevailing wind direction was the southeast where it is the direction of the sea coast from the city (de Miranda et al., 2002). However, we also found a weak correlation between Na and Cl, this result might be explained by a considerable amount of Cl particle converted to gas-phase HCl by the reactions with HNO_3 and H_2SO_4 in polluted urban atmosphere like São Paulo City (Kang et al., 2014). Group 4 is characterized with K and P, which explains 10% of the variance and may be related with biomass burning by mainly biofuel usage in the city. Andrade et al. (2012) linked the biofuel usage with the K emission in Sao Paulo City study. Group 5 is characterized with Mg and S, which explains 9.83% of the variance and may be related with fossil fuel combustion.

Our findings in this study indicate the possibility of tree bark as a suitable passive biomonitor to estimate the spatial distribution of air pollution exposure. Generally, the urbanization in developing countries occurs without planning the land use and without considering the impact on human health. Therefore, the employment of this methodology in order to estimate the spatial distribution of air pollution may play an important role to improve the planning of urbanization. The associations between increased levels of air pollution and risks of mortality and morbidity have been studies for a long time and is already well-established (Pope et al., 2009; Kampa and Castanas, 2008; WHO, 2013). Recent epidemiological studies in urban areas suggest that chronic health effects of air pollution exposure may be even three times higher than previously reported (Jerrett et al., 2005).

The epidemiological studies have used conventional air pollution network from monitoring stations, which were unable to determine the accurate intra urban spatial variation of air pollutants and to access the accurate health effect with minimizing the error in exposure risk assessment. We suggest a potential approach to overcome this drawback using the chemical analysis of tree bark samples which are collected at various sites with a higher spatial resolution in the study area. However, note that these are difficult to employ for a large area due to the higher cost and the complexity of logistics execution. Currently, some studies have employed the spatiotemporal land-use regression models to improve the consistency of association between air pollution exposure and human health effects (Wang et al., 2013; Yang et al., 2015). Combing with the land-use regression model, our approach could be a useful alternative and provide a higher spatial resolution of air pollution to access accurate health effect and risk assessment of air pollution.

5. Conclusion

Our results identified the areas impacted by more abundant trace elements, and indicated specific plausible sources using a biomonitoring technique in the intra urban area of São Paulo City. The biomonitoring technique by determining the elemental composition accumulated in tree barks can be useful to identify major sources of air pollution in urban atmospheres. Our results can be used for possible new street classification based on vehicular emissions. These results show that it is possible to differentiate particulate matter from one point to another, even within a small area, as well as sources of vehicular emissions.

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