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Distribution of Trace Metals in Street Dusts and Tree Leaves and Their Source Identification in a Mid-Populated Anatolian City

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Abstract

Sivas is a central Anatolian city in Turkey with mid-population. Due to its geographical structure high air pollution episodes can be observed in the atmosphere of Sivas. A study with city wide sampling campaign can help to identify the possible sources. Therefore, we aimed to investigate the multi-elemental and multi-point analysis of trace metals in street dusts and tree leaves in Sivas. In the street dusts, Ca concentration was $>10\%$. Fe, Cl, and K contribution was 2.5%, 1.75%, and 1.1%, respectively. The average S and Ti concentrations were between 1 and 10 mg/g, I, Mn, Sr, Cr, V, Ba, Zn, Ni, Zr, and Cu were between 1 and 0.1 mg/g, and W, Pb, Sn, Th, Rb, Sb, Co, Bi, As, U were between 0.1 and 0.01 mg/g, in the ascending order. The same elements were investigated for vegetation. Ca, K, S, and Cl were the elements with highest contribution.

Keywords Street dust · Leaf · Trace metal · Source identification · PCA

Cities are intensive residential area with dense anthropogenic activities related to urbanization and industrialization such as construction, population, transportation, production, consumption, and waste compose, etc. Due to heavy urbanization and industrialization, nearly half of the population in the world now lives in urban areas. The dense urbanization and industrialization cause an increasing amount of toxic contaminants being discharged to urban environment. As a result of this discharge, a variety of environmental problems have emerged. Trace metal pollution is the principal problem, especially in downtown areas (Kumar et al. 2015). Airborne particulate matter is one of the major factors affecting atmospheric environmental quality. It was showed that particulate matter (PM) carried many organic (Kuzu 2019), inorganic toxic substances (Kuzu et al. 2013), and harmful microorganisms (Safatov et al. 2008) that have potentially detrimental effects on the environment and human health.

The term "trace metals" refers to any metallic element that has a relatively high density and is toxic or poisonous even at low concentrations. Trace metals are named after the group of metals and metalloids with atomic density greater than 4 g/cm^3 or 5 times or more, greater than water (Duruibe et al. 2007). Trace metals include Si, Ca, Al, Fe, K, Na, Ba, P, Mg, Mo, Be, Co, Mn, Ni, Pb, Cd, Cu, Ti, Zn, Cl, Cr, B, S, Hg, As, Ag, Zr, Sn, Bi, Ga, Sb, Br, La, Rb, In, Nb, Nd, Eu, Sm, Tb, Th, Lu, W, Se, V, Pu, Sr, U, Sc, Sb and etc. (Nuhoglu and Bülbül 2003).

Street dust is one of the important air pollution indicators that reflect the status of traffic, urbanization, industrialization, and natural soil.It is also an important pathway because it usually carries toxic trace metals. Airborne PM are deposited on vegetation, roofs, garden, and road. Usually street dusts and other emissions from combustion sources may have harmful effects on human health because it contains toxic substances. PM, containing trace metals, can possibly enter the human body through the respiratory system and cause bioaccumulation, which can lead to acute and chronic poisoning.

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Epidemiological studies have shown the evidence of adverse effects of PM pollution (Dockery and Pope 1994).Many trace metals have been identified as having adverse public health effects due to their toxicity. Several studies revealed that human exposure to high concentrations of trace metals may lead to bioaccumulation in the fatty tissues of the human body, disrupt the normal functioning of the internal organs (Bocca et al. 2004), affect the central nervous system (Govoni et al. 1988), and may be deposited in the circulatory system (Waisberg et al. 2003).

Many studies showed that children exposure to contaminated soils, dust, and air particles may cause ingestion of significant amounts of toxic elements through the hand–mouth pathway and through other routes of exposure (Mielke et al. 1999; Raghunath et al. 1999). Among these metals, Cadmium (Cd) is well known as a global contaminant (Tchounwou et al. 2012). Cadmium has become a threat to human health when it passes up the food chain (Satarug et al. 2003). Cd has toxic effects on microorganisms, plants, animals, and humans. There is sufficient evidence in humans for the carcinogenicity of Cd and Cd compounds and for genotoxic effects of its ionic forms in a variety of types of eukaryotic cells. Cd enters the body mainly by inhalation and by ingestion (Waisberg et al. 2003). Cd is a significant example of the components on particles. An extensive list of metals in the environment and their toxic effects were listed elsewhere (Tchounwou et al. 2012).

The trace elements can deposit and accumulate at different parts of vegetation. Air is an important pathway of the pollutants that deposit on vegetation (Kosiorek et al. 2016). Pine needles are reported to be good indicators of air pollution (Holoubek et al. 2000). Climatic conditions and age of the tree are also important factors that affect the needle metal concentration (Varnagirytė-Kabašinskienė et al. 2014). Many researchers have studied trace metal concentrations on plants (Yilmaz and Zengin 2004; Parzych et al. 2017; Cindrić et al. 2019). The results varied over a wide range due to temporal and geographical differences between the studies.

There are many studies of trace metals contamination of street dust in urban areas; however, little attention has been paid to determine those contamination at different environmental matrices and determine their possible sources. This research aims to (a) determine the content of toxic elements in street dust and tree leaves, (b) investigate the relation between to traffic and domestic fuel through enrichment factor and principal component analysis.

Materials and Methods

Sivas is a central Anatolian city, where lies over a vast plain (between 38° 32′ N and 40° 16′ N latitudes and 35° 50′ E and 38° 14′ E longitudes). The total urban area of Sivas is 115.50 km^2, and its population is 359,219 and the number of registered motor vehicle are 135,000 of which 52.1% is personal cars, 2.2% minibuses, 0.8% buses, 16.1% vans/pickups, 3.9% trucks, 4.3% motorbikes, 20.3% tractors, and 0.3% vehicles for special purposes, at the time of sampling (TSI 2019). The major industries are cement factory, railway car industry, iron and steel factory (23 km to the south), tile and brick factories, molding factory, automotive spare parts, cotton spinning mill and carpet weaving workshops. The major climate systems of Central Anatolia is Bsk (Kuzu and Cetinkaya 2019) and Dsb (Türkeş 2010) according to Köppen-Geiger climate systems. Sivas has snowy forest climate with dry summer, a typical continental-cold climate, which is within Dsb classification.

For dust sampling, the study area was divided into two research sites: commercial-residential and control (background) areas. The control site was within Sivas Cumhuriyet University Campus, where is 6–7 km away from the Sivas city center. The traffic density at the control site is much lower than in Sivas city. The commercial-residential site is mostly located in the center of city and consists of dwelling house, shops, offices, workshops, petrol pump, motor garage, welding shop, and etc.

Street dust samples were collected in January and August 2015 from various locations of Sivas. The selection of the sampling points was based on to represent different characteristics of the city. Ten different sampling points were chosen in order to determine the distribution of trace metals in street dusts and tree leaves. The location of the sampling points within the city are shown in Fig. 1.

The sampling point shown as 9 (the most southern location) is the background sampling location. In the sampling zones, we aimed to take samples from the scots pine trees. Scots pine tree was not present at all of the sampling locations. Therefore, we took samples from existing trees, where scots pine was not available. For this reason, the 1, 6 and 8 research areas in the scots pine, 2, 3 and 4 in the research area of spruce, 5, 7, 9 and 10 of the research area were taken from the thuja trees. The young shoot samples, taken from the hand-reaching height of the plants (average height from 1.5 to 1.8 m), were brought to the laboratory and dried in an oven at 105 °C for one day and shredded in grinder as thin powder. Afterwards, the shredded samples were analyzed by X-ray fluorescence (XRF) analyser.

Outside the urban area, huge arid texture is present with sparse vegetation. Therefore, meteorological parameters can influence mechanical dust formation. Furthermore, Anatolian Peninsula is prone to dust transportation from proximate desert areas such as Arabian Peninsula. Air masses from desert transportation reach to the area from southeast direction. Windrose is prepared from METAR data of Sivas Airport. The windrose is given in Supplementary Material Fig. 1. The dominant wind direction is the North-northwest

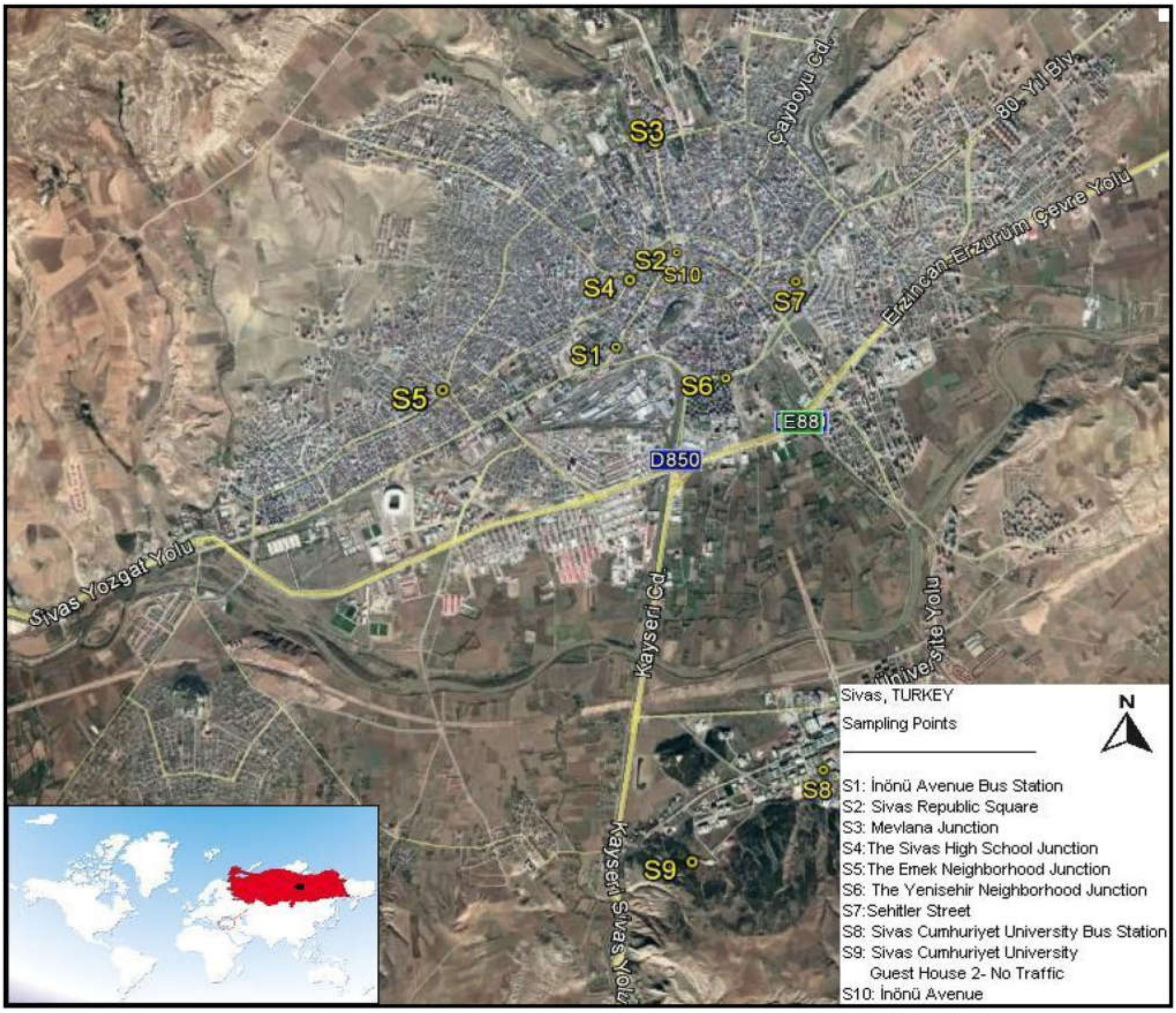

Fig. 1 Sampling locations across Sivas city

direction. South-easterly winds are the second dominant wind direction for the sampling area.

Street dust samples were collected from a 2 m^2 area on the edge of the highway (so as to characterize the entire city) by sweeping with a clean plastic dustpan and brush. The collected samples were placed in a clean polyethylene bag and transferred to the laboratory. The solid particles in the samples like stones and gravels are removed though a sieve, which had 2 mm side length. All the samples were dried in an oven at 105 °C for 24 h as with vegetation samples. The sieved samples were analyzed by Energy Dispersive X-ray Fluorescence (EDXRF) spectrometry.

EDXRF is one of two general types of X-ray Fluorescence techniques used for elemental analysis applications. All of the elements in the sample are excited simultaneously in EDXRF spectrometers and an energy dispersive detector in combination with a multi-channel analyzer is used simultaneously to collect the fluorescence radiation emitted from the sample and then separate the different energies of the characteristic radiation from each of the different sample elements. EDXRF is a three-axial energy-dispersive spectrometer, with a conventional silver X-ray tube and a molybdenum secondary target arrangement. This geometry has been shown to have the best peak-to background conditions. The design was arranged in such a way that the sample is positioned in the horizontal plane. The radiation from the sample was detected by an Si(Li) detector (active area 80 mm^2, FWHM at 5.9 keV of 173 eV). The X-ray tube was operated at a voltage of 55 kV and a current of 25 mA. The life time of each spectrum was 1000 s. An X-MET8000 handheld

XRF analyzer was used in this study. The analyser can detect and quantify elements heavier than Ti and lighter than U. The combination of a high performance X-ray tube and large area silicon-drift detector (SDD) delivers the performance required for even the most demanding QA/QC applications. The device is pre-calibrated and does not require calibration before analyses. In this study, QA/ACRM were not measured specifically.

Enrichment factor (EF) was employed to determine the degree of contamination from anthropogenic sources. Enrichment factor is calculated according to Eq. 1.

$$EF = \frac{C_n/C_{ref}}{B_n/B_{ref}} \tag{1}$$

where, C_n is the concentration of an element "n" in the environmental sample. C_{ref} is the concentration of reference element in the sample. B_n is the concentration of element "n" is the Earth's crust and B_{ref} is the concentration of reference element in the Earth's crust. the common reference element is aluminium. However, in the absence of this element, another components can be selected (Anil et al. 2019). In this study, we used Fe as reference element for EF calculations.

Results and Discussion

This study investigated the elemental composition of street dust and leaves in Sivas city, where light industrial activities and traffic strongly affect trace metal distribution. The analysis results of road dust samples are given in Supplementary Material Table 1(a) and (b).

We investigated Ca, Fe, K, Ti, S, Cl, Sr, Mn, Ba, V, Zr, Ni, Zn, Cr, Cu, Co, Pb, As, Se, Rb, Sn, Th, P, W, Bi, Hg, U, Mo, Sb, Cd, Ag, and I components. As being an earth element Ca had the highest concentration. Its amount was > 10% at all sampling locations except sampling no 1 in the second sampling campaign. Sampling no 1 is located near a bus stop in one of the most crowded streets of the city. Fe and Cl was the second and third highest contributors to the street dust samples. Fe amount in the dust samples varied between 0.02% and 5.3%. Its average value was 2.5%. Cl amount ranged from ND to 3.4% with an average of 1.75%. In the first sampling, Cl amount in the dust samples were lower than K. However, Cl had higher amount in the second sampling. The average of K was 1.1%, ranging between 0.3% and 2%. The average S and Ti concentrations were 8.9 and 2.4 mg/g, respectively. The average concentrations of I, Mn, Sr, Cr, V, Ba, Zn, Ni, Zr, and Cu were between 1 and 0.1 mg/g, in the ascending order. The average concentrations of W, Pb, Sn, Th, Rb, Sb, Co, Bi, As, U were between 0.1 and 0.01 mg/g, in the ascending order. Mo and Se had concentrations lower than 0.01 mg/g. P, Hg, Cd, Ag was not detected in any of the samples. Results of this study showed similarities with some results present elsewhere. Valotto et al. (2019) analysed road dust samples for major and trace elements in Italy. As with our study, Ca was > 10%. Fe was the second highest contributor with 1.7%. Remaining compounds were comparable within an order of magnitude. Gunawardana et al. (2012) determined the composition of road dusts at Australia's growing urban regions. Cr was two order of magnitude, Fe was four orders of magnitude higher in Sivas than in Australia. The abundance of Ti and Fe rich particles are attributed to coal combustion and other metallurgical emissions (Senlin et al. 2008). Sezgin et al. (2004) collected road dust samples in Istanbul, where the most crowded city of Turkey. Pb concentrations were higher in Istanbul, whereas Ni concentrations were higher in the dust samples in Sivas.

Temporal differences did not show much variance for most of the elements. The highest variance was for Cl. This is the tracer of marine aerosols. However, Sivas is far from any coastline of Turkey. Cu and Pb had significant difference between 1st and 2nd sampling campaign. Both elements are the tracers of traffic related emissions (Jin et al. 2019). Cu had the highest concentration at sampling no 2, where is the centre of the city. High Cr concentrations were observed during the first sampling campaign at sampling locations 8 and 9. Those locations correspond to the university campus, located to the south. This elevated concentrations indicated transportation from industrial applications to background sampling site.

In order to compute the degree of contamination from anthropogenic sources EF was calculated for each element. EF ratio higher than 10 points out anthropogenic influence on the measured values (Summak et al. 2018). We showed the results of EF in Fig. 2. S, Cl, Sn, Th, As, and Se had significant enrichment, being > 10. Among those components, Cl and Se had EF > 100. Elevated Cl was observed in summer sampling. Atmospheric Cl has natural sources including wild fires and dust storms, as well as anthropogenic sources, such as coal combustion, biomass burning, industrial emissions, and road salt application (Luo et al. 2019). Dust storms, arising from desert areas, has a strong influence in Anatolia due to being close to countries such as Syria and Iraq (Acar and Demiryürek 2019). Therefore, dust storms can be the natural source of Cl burden on dust particles. On the other hand, many combustion related activities may be the anthropogenic source of Cl. Especially, S is an important component of sulphur containing fuel combustion. The highest S concentration was observed at the S10 during both seasons. Although the highest concentration was in winter, the average of summer sampling was higher than the average of winter sampling. This indicates the resuspension

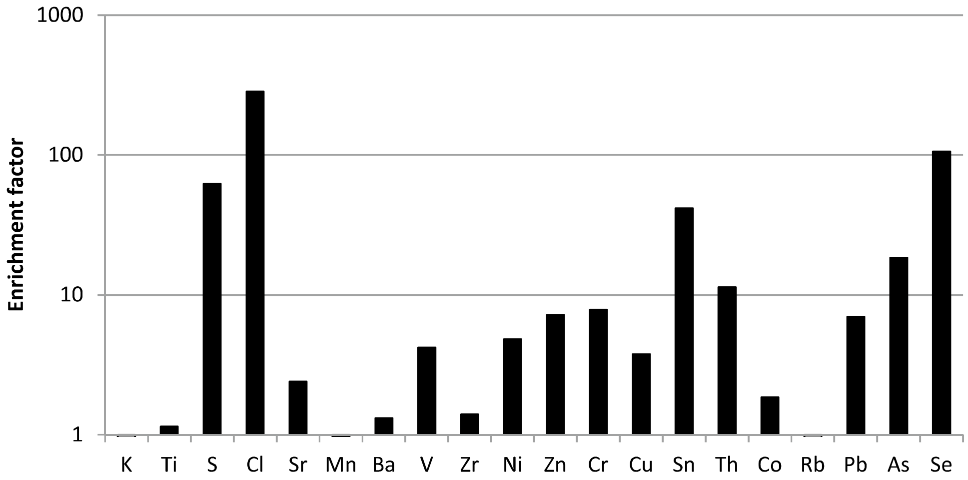

Fig. 2 Enrichment factor of street dust

Table 1 Varimax rotated component matrix of dust samples (N = 17)

	Component				
	1	2	3	4	5
Ca	– .914	.178	– .110	.021	.311
Fe	.258	– .080	.857	.040	– .085
K	.759	– .182	.401	– .011	– .233
Ti	.948	.063	.055	.076	– .114
S	– .119	.860	– .109	– .071	.200
Cl	– .068	.636	– .398	– .524	.174
Sr	– .602	.467	– .275	– .146	.418
Mn	.649	– .201	.529	.160	.315
Ba	.858	– .127	.237	.133	– .345
V	– .032	– .013	.150	.900	.007
Zr	.895	– .149	.140	– .094	.058
Ni	– .400	.184	– .182	– .028	.853
Zn	– .254	.810	– .175	– .175	– .194
Cr	.083	– .224	– .010	.010	.838
Cu	– .113	.278	.033	– .895	– .078
Sn	– .148	.464	– .520	– .342	– .098
Th	.296	– .075	– .138	– .036	– .596
Co	.902	.297	.060	.127	– .045
Rb	.907	– .281	.226	– .062	– .068
Pb	.043	.868	.134	.071	– .071
As	.298	.373	.613	.466	– .111
Se	– .394	.225	– .578	.498	– .152

effect in summer months. Weathering of Se-including rocks and coal combustion are primary sources of Se (El-Ramady et al. 2015).

We employed principal component analysis (PCA) in order to reduce dimensions of the list and sort them as groups. Varimax rotation was applied and variables with factor loadings greater than 0.50 were chosen to interpret element member at each group. Results of the dust sample PCA analysis is given in Table 1.

First component was comprised of Rb, Co, K, Ti, Mn, Ba, and Zr. This component explained 31.6% of the total variance. Source of those elements likely to demonstrate natural soil contribution (Wang et al. 2016) but they cannot be limited to soil contribution alone. Second component comprised of S, Pb, Sn, and Zn. It explained 16.7% of the total variance. It showed contribution from traffic source (Shi and Lu 2018; Zhang et al. 2019). Third component was consisted of Fe, Mn, and As with explaining 12.1% of the whole variance. The third component indicated soil parent materials/geogenic sources (Jin et al. 2019). Forth component was comprised of V and Se. This component explained 11.9% of the total variance. Selenium is reported to be present in high-sulphur coals (Yudovich and Ketris 2006). Zhang et al. (2011) expressed that vanadium is a common element in stone coal. Therefore, the forth component can attributed to coal combustion. The fifth component was comprised of Cr and Ni. It explained 11.5% of the total variance. It shows typical emissions from alloying (Nishiyama et al. 2006) or welding activities. Those five components could explain the 83.7% of the entire composition. The predominant anthropogenic contributor was traffic. Almost half of the dust were originated from natural origin. Together with EF results, PCA results show significant contribution from natural origin such as dust transportation.

Vegetation component can act as an indicator of anthropogenic emissions. Therefore, spatial differences in their composition has a potential in describing elemental inputs to the vegetation. The analysis results of leaf samples are given in Supplementary Material Table 2(a) and (b). As with dust composition, the highest contributor was Ca. Its average amount was 2.24%. K, S, and Cl had average concentrations of 9.36, 4.00, and 1.86 g/kg, respectively. The average Fe, Mn, and Ti concentrations were 1.00 0.10, and 0.10 g/kg, respectively. I was only observed in one sample with 0.11 g/kg concentration value. The average concentrations of Sr, Zn, Ba, Cr, Ag, and Cd were 89, 54, 34, 15, 15, and

Table 2 Varimax rotated component matrix of needle samples (N = 20)

	Component				
	1	2	3	4	5
Ca	.546	− .432	.214	− .089	.359
Fe	.966	.051	− .180	− .037	.056
K	− .115	.330	.651	.357	.353
Ti	.666	− .554	.036	− .380	− .088
S	.761	− .418	.230	.044	.014
Cl	.825	− .366	.170	− .016	− .056
Sr	.206	− .433	.554	.287	.132
Mn	.730	.575	.156	.026	.054
Ba	.380	.713	.108	− .482	.171
Zr	.747	.228	− .399	.187	.180
Zn	.748	.214	− .413	.051	− .127
Cr	.747	.409	.327	.002	− .143
Rb	.258	.589	− .061	.641	− .149
Pb	.895	− .059	− .121	.010	.055
Cd	− .217	.011	− .372	− .015	.873
Ag	− .198	.607	.357	− .546	.004

12 mg/g, respectively. Cu, Zr, As, and Co concentrations were 20, 3.3, 2.6, and 2.1 mg/kg, respectively. But these concentrations were encountered only once during the entire sampling. V, Ni, Sn, Th, Se, P, W, Bi, Hg, U, Mo, and Sb were not detected in any of the samples. Ca, K, Fe, Ti, S, Cl, Sr, and Mn was the major elements in the pine needles. Ba, Zn, Cr, Rb, Pb, Cd, Ag, Zr, Cu, Co, and As was less significant metal components in the needles. Those major and minor elements represent typical pine composition (Pongrac et al. 2019). Ca, K, S, and Sr concentrations were higher in thuja, whereas Fe, Mn, Zn, and Cr were higher in scots pine. The lowest Ti and Cl concentrations were observed on spruce samples. It is well known that element composition of leaf samples can be enriched by anthropogenic influence (Sulaiman and Hamzah 2018). In order to determine the anthropogenic contribution we also employed PCA to the analysis results of needle samples. Results of the PCA analysis is given in Table 2.

First component was comprised of Ca, Fe, Ti, S, Cl, Mn, Zr, Cr, and Pb. It explained 27.7% of the total variance, whereas the second component was comprised of Mn, Ba, Rb, Ag. 25.2% of the total variance was explained by the second component. The third component was K and Sr, which explained the 12.6% of the total variance. Those three components exhibited typical elements of plant composition (Johansson 1995; Baath 2008; Aniszewska et al. 2017). The fourth component was Rb alone with explaining 9.8% of the total variance and the fifth component was Cd alone with explaining 8.3% of the total variance. Component 4 was present in similar quantities at all trees. The similar spatial and temporal pattern indicated that it has to be an essential component of the tree composition. Component 5 was comprised of Cd, which is a well-known anthropogenic pollutant. Cd was not present in all samples but it was available during both sampling campaigns at city centre and background sampling site.

Increased attention has been paid to trace metal distribution on environmental matrices over the several decades. The distribution of the environmental contaminants and their EF and PCA analysis provides essential information of possible sources. Therefore, such studies has been conducted at different environmental compartments in order to determine their variability and levels. In this study, EF results showed that street dust samples were significantly enriched with Cl, Se, S, Sn, As, and Th. Origin of those elements can either be natural or anthropogenic. Latter analysis was performed with PCA. Five factors were generated for street dust samples. The results indicated that the principal source of trace metals were motor vehicle, burning of domestic fuel, and natural dust contribution. Road dust did not provide a significant differentiation between the sampling points. There were only some minor spatial and temporal differences. The same applies for the vegetation composition. This indicated that the air pollutants are distributed throughout the city.

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