

Active moss biomonitoring of small-scale spatial distribution of airborne major and trace elements in the Belgrade urban area

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Abstract In urban environments, human exposure to air pollutants is expected to be significantly increased, especially near busy traffic streets, street canyons, tunnels, etc. where urban topography and microclimate may additionally cause poor air conditions giving rise to pollution hotspots. As a practical and cost-effective approach, active moss biomonitoring survey of some major and trace element air pollution was performed in the Belgrade street canyons and city tunnel in 2011 with the aim to evaluate possibility of using *Sphagnum girgensohnii* moss bags for investigation of the small-scale vertical and horizontal distribution patterns of the elements. In five street canyons, the moss bags were hung at heights of about 4, 8 and 16 m, during 10 weeks, and also, for the same time, the moss bags were exposed in the tunnel, in front of and out of it. After the exposure period, the concentrations of Al, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sr, V and Zn in the moss were determined by inductively coupled plasma optical emission spectrometry. According to the results, in all street canyons,

the vertical distribution patterns of the moss elements concentration (Al, Ba, Co, Cr, Cu, Ni, Pb, Sr, V and Zn) showed statistically significant decrease from the first to the third heights of bags exposure. In the tunnel experiment, from inner to out of the tunnel, for Al, Ba, Cd, Co, Cr, Cu, Fe, K and Zn, decreasing trend of concentrations was obtained. Significantly higher concentration of the elements was pronounced for the tunnel in comparison with the street canyons. The results indicate that the use of *S. girgensohnii* moss bags is a simple, sensitive and inexpensive way to monitor the small-scale inner city spatial distribution of airborne major and trace element content.

Keywords Active biomonitoring · Major and trace elements · Moss bags · *Sphagnum girgensohnii* · Urban area · Spatial distribution

Introduction

It is well recognised that many substances emitted by vehicles (PMs, toxic elements) represent a hazard for the human health (Fenger 1999; WHO 2005), leading to human exposure to toxic substances in urban area for both pedestrians and residents being generally high (Colville et al. 2001). Several epidemiological studies have reported links between particulate air pollution, as well as trace elements associated with PM and health problems (Seaton et al. 1995; Harrison and Yin 2000).

Different trace elements can behave differently with regard to their vertical distribution pattern, depending on their own nature, their main form of diffusion (e.g. in association

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with coarse or fine airborne particles), their primary source, etc. The concern for the vertical distribution pattern of trace elements in the urban environment is rapidly growing. Human exposure to hazardous substances is expected to be significantly increased, especially in parts near a busy traffic axis in the city centre, where urban topography and microclimate may contribute to the creation of poor air dispersion conditions, giving rise to contamination hotspots (Vardoulakis et al. 2003). High pollution levels have been observed in microenvironments such as street canyons, city tunnels, etc. The term street canyon ideally refers to a relatively narrow street with buildings lined up continuously along both sides (Nicholson 1975). However, the same term has been used to refer to larger streets, also called avenue canyons. In the real world, a broader definition of the term has been applied, including urban streets that are not necessarily flanked by buildings continuously on both sides, allowing thus for some openings on the walls of the canyon. Within these streets, pedestrians, cyclists, drivers and residents are likely to be exposed to pollutant concentrations exceeding current air quality standards (Vardoulakis et al. 2003). Furthermore, city tunnels have been built to facilitate mobility of people within the cities. An efficient tunnel construction has the potential to reduce traffic congestion. On the other hand, since ventilation outlets collect vehicle exhausts and release it all in one or two locations, road tunnels in urban areas give rise to problems including localised air pollution (Ma et al. 2004).

In general, monitoring stations and/or samplers should be located near places of expected air pollution hotspots. Also, their selection must be reasonable, reflecting population exposure over the averaging times. The total number of air quality monitoring stations or sampling locations within a city is limited by practical constraints. However, biomonitoring can give a reliable picture of pollution patterns in a much more cost-effective way and can also integrate over a longer period than most of physical measurements do. A biomonitor is an organism (or part of an organism or a community of organisms) that contains information on the quantitative aspects of the quality of the environment (e.g. Markert et al. 2003). Given their morphological and physiological characteristics, mosses have proved to be suitable biomonitors for a wide range of pollutants. Lacking a root system or cuticle, they absorb water, nutrients and toxic substances mainly via the entire plant surface from dry and wet deposition (Brown and Bates 1990). Mosses show a resistance to various toxic compounds (e.g. some trace elements), which accumulate in moss tissue and can therefore be used for their monitoring. A review of applications in the broad field of trace element monitoring by mosses has been given in Zechmeister et al. (2003). Most applications were in the field of passive biomonitoring, where mosses were taken from their natural growth sites (e.g. UNECE ICP Vegetation Report 2008). However, this international programme is restricted to rural sites only, and there are

only a very few data (e.g. Zechmeister et al. 2006) on urban environments obtained by this method.

Active biomonitoring, using moss as transplants, predominantly in urban and industrial areas where naturally growing mosses are usually absent, has been studied as a flexible and low-cost method to achieve information about trace elements pollution. Since the early work of Little and Martin (1974), moss bags have been mainly used in terrestrial and aquatic ecosystems to assess industrial (Fernández et al. 2000, 2004; Carballeira et al. 2006), urban (Adamo et al. 2003, 2007; Giordano et al. 2005; Culicov et al. 2005; Aničić et al. 2009a), indoor (Al-Radady et al. 1993) and road traffic pollution (Zechmeister et al. 2006). In biomonitoring studies, spatial deposition patterns of trace elements are generally regarded as being only horizontal, while vertical distributions are also important, especially in the so-called breathing zone of the urban areas, where they can affect the human health. The breathing zone of an urban area includes all heights within the surface layer of street canyons at which people act and breathe.

In this study, active moss biomonitoring survey of trace element air pollution in the Belgrade urban area was performed. The main aim was to evaluate possibilities of using *Sphagnum girgensohnii* moss bags for investigation of small-scale inner city vertical and horizontal distribution patterns of selected major and trace elements in the street canyons and the city tunnel. Firstly, since the traffic is a major source of pollution in the selected street canyons, we hypothesised that concentration of determined elements would decrease with increasing height. Secondly, assuming that one of the streets is a pedestrian zone, we supposed that concentration of traffic-related metals would be lower in this street. Thirdly, in the tunnel experiment, decreasing trend of element concentrations from inside to outside of the tunnel was expected. Also, the time-dependent accumulative property of *S. girgensohnii* was assessed in this experiment. Finally, the factors influencing typical spatial distribution were considered.

Material and methods

Moss bags preparation

Moss *S. girgensohnii* Rusow was collected at the end of May 2011 from a pristine wetland area located near Dubna, Russian Federation. This background area was chosen on the basis of results obtained in previous studies (Aničić et al. 2009b, c). In the laboratory, green upper part of moss was separated and just manually carefully cleaned from soil particles and other foreign matter. Moss material was air-dried and gently hand mixed to obtain homogeneous sample. About 3 g of the moss was packed loosely in 10×10 cm nylon net bags with 2-mm mesh size. To avoid the moss

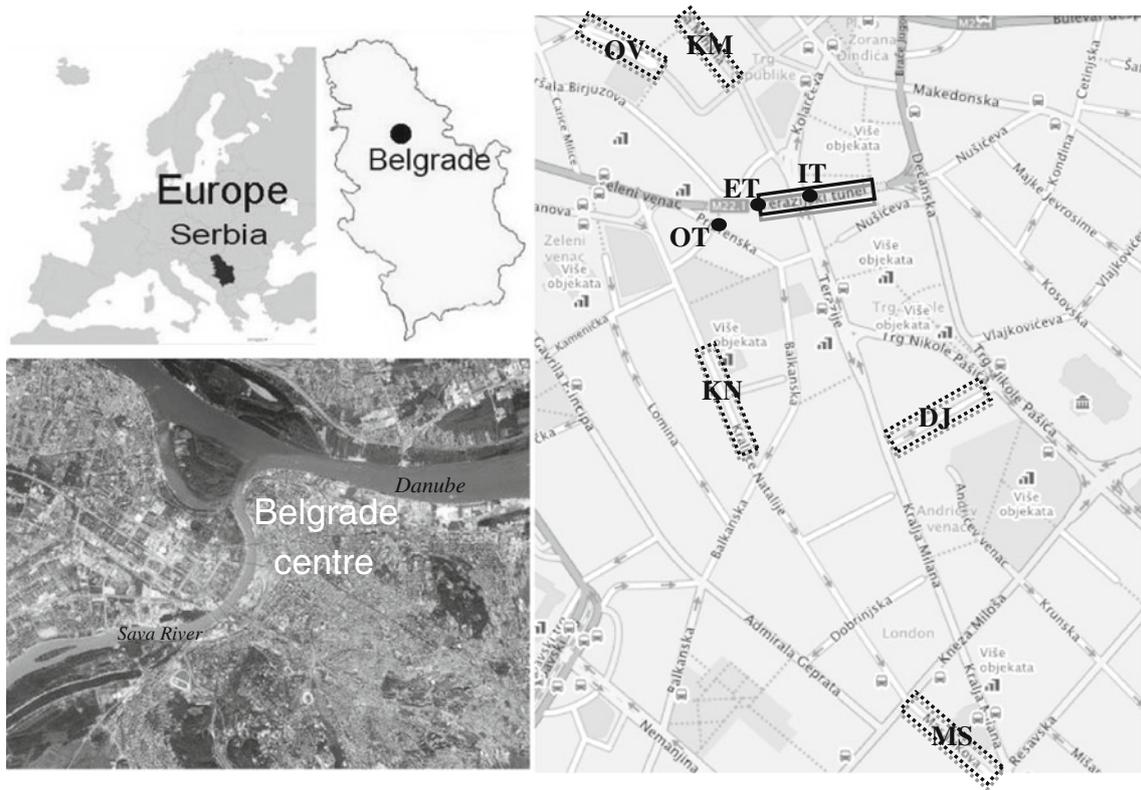


Fig. 1 Map of the studied sites in the Belgrade urban area: street canyons (KM, OV, DJ, KN and MS) and city tunnel (IT, ET and OT)

material contamination, sampling and preparation of moss bags were carried out wearing polyethylene gloves.

Experimental set-up

The experiments were performed in five street canyons and one city tunnel situated in heavily traffic area of the Belgrade City ($\varphi=44^{\circ}49'N$, $\lambda = 20^{\circ}27'E$, $H_s=117$ m), the capital of the Republic of Serbia (Fig. 1). The traffic flow measured in the street canyons were in a range of 212 to 1,254 vehicles per hour, as indicated by counting at the morning rush hour during the experiment. The tunnel, with an average traffic intensity of 1,451 vehicles per hour, can also be considered as a traffic intensive road. Outside of the tunnel, in Prizrenska Street,

average traffic intensity was 1,451 vehicles per hour (City Secretariat for Transport 2011). Additionally, there are large multi-level public garages in four of the selected street canyons (KN, MS, DJ, OV), while the fifth street (KM) is a pedestrian zone.

Following the geometry rules described by Vardoulakis et al. (2003), according to height/width (H/W), as well as length/height ratio (L/H), the chosen street canyons can be classified as regular, medium or short and rather symmetric (Table 1).

There is uniform set-up in all street canyons: the moss bag holders were mounted on the buildings at a distance about 25–30 m from the garage entrance. Moss bags were hung at heights of about 4, 8 and 16 m, and referred as z/H ratio in

Table 1 Dimensions and types of the studied street canyons in the Belgrade urban area

Street name	Street dimensions [m]			H/W	Type	L/H	Type	Street symmetry
	Height (H)	Width (W)	Length (L)					
1. St. Kraljice Natalije (KN)	27	20	160	1.35	Regular	5.93	Medium–long	Symmetric
2. Masarikova St. (MS)	25	16	140	1.56	Regular–deep	5.60	Medium–long	Asymmetric
3. St. Dragoslava Jovanovića (DJ)	25	9	50	2.78	Deep	2.00	Short	Symmetric
4. Obilićev venac (OV)	28	22	80	1.27	Regular	2.86	Short	Symmetric
5. St. Kneza Mihaila (KM)	25	15	500	1.67	Regular–deep	20.00	Long	Symmetric

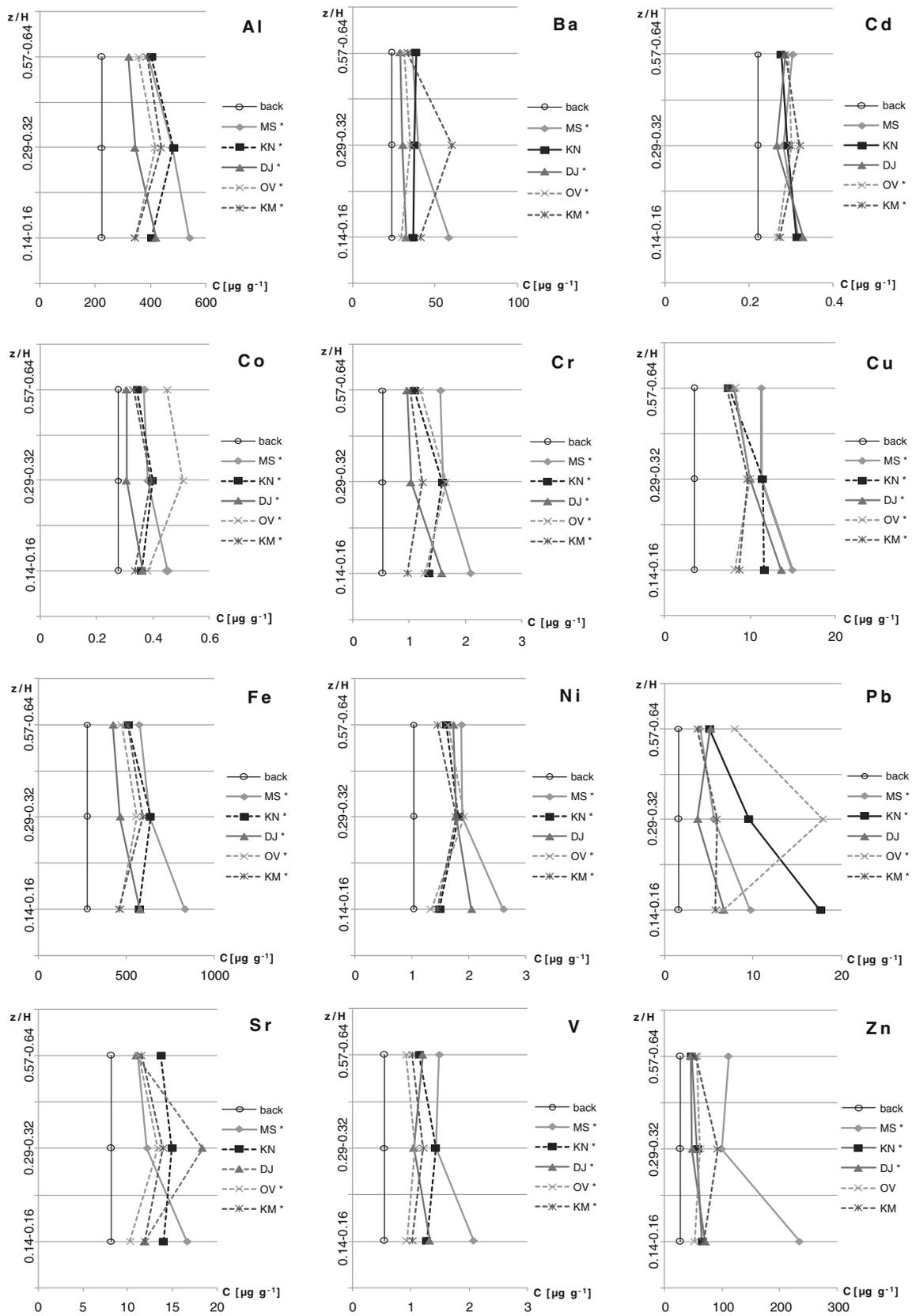


Fig. 2 Vertical profiles of element (Al, Ba, Cd, Co, Cr, Cu, Fe, Ni, Pb, Sr, V and Zn) concentrations ($\mu\text{g g}^{-1}$) in the moss bags exposed for 10 weeks in five canyon streets (MS, KN, DJ, OV and KM), and the initial element

concentrations in the unexposed moss (back); z/H , heights of about 4, 8 and 16 m above the ground; $*p < 0.05$, statistically significant difference between heights of moss bags exposure

Table 2 Relative accumulation factors (RAF) of the elements in moss samples after 10 weeks of exposure in five canyon streets in Belgrade urban area

RAF	Element
≤0	K, Mn, Mg
0–1	Ca, Cd, Co, Na, Sr
1–5	Al, Ba, Cu, Fe, Ni, V
5–10	Cr, Pb, Zn

all street canyons (0.14–0.16, 0.29–0.32 and 0.57–0.64, respectively), at a distance of about 1–2 m of the wall of the building, away from any porches, balconies, etc. The first height was chosen in order to gain an insight into the pollution level in pedestrian zone and also to prevent vandalism removal. The other two heights were selected arbitrary because, according to our knowledge, there is no standardised protocol prescribing selection of sites for the study of vertical distribution of trace elements in urban environment.

In the city tunnel experiment, the bags were exposed inside (IT), in front of the entrance (ET) and outside of the tunnel (OT) in order to test horizontal element distribution in the moss.

The moss bags in both experiments were exposed for 10 weeks (selected on the basis of the previous study, Aničić et al. 2009c) during the summer–autumn of 2011. In the tunnel experiment, the half of bags was removed from the exposure sites after 5 weeks searching for time-dependent of element accumulation by the moss.

Chemical analysis

After the exposure periods, in the laboratory, approximately 0.3 g of each air-dried homogenised moss sample was digested for 45 min in a microwave digester (MILESTONE ETHOS 1) with 7 ml of 65 % HNO₃ (Sigma Aldrich) and 1 ml of 30 % H₂O₂ (Sigma Aldrich) at 200 °C. Digested samples were diluted with distilled water to a total volume of 50 ml. The concentrations of 17 elements (Al, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sr, V and Zn) in the moss were determined by inductively coupled plasma optical emission spectrometry (ThermoCAP6500 duo). For calibration, multi-element stock solution (Merck) containing 1.000 µgml⁻¹ of each determined element was used to prepare intermediate multi-element standard solutions: 50, 20, 1, 0.1, 0.01 and 0.005 µgml⁻¹. The quality control was performed using the standard reference material lichen-336 (IAEA).

Data analysis

The data were processed using StatSoft STATISTICA 6.0. Basic, non-parametric statistics (Mann–Whitney *U* test) was used to check for significance of differences at *p*<0.05 level in all cases, and correlation between elements were calculated.

In order to assess the element accumulation of *S. gigensohnii*, relative accumulation factors (RAF) were

calculated as the mass content of each element after exposure (*C*_{exposed}) reduced and divided by the element content before exposure (*C*_{initial}):

$$RAF = (C_{exposed} - C_{initial}) / C_{initial}$$

Results and discussion

Vertical element distribution in moss bags—street canyon experiment

Concentrations of most elements were statistically higher in the exposed moss samples than in the background ones (initial moss element concentrations), indicating that the street canyons in which the moss bags were exposed were polluted by the elements in question (Fig. 2). According to calculated RAF values, the most accumulated element in moss bags was Pb, then in descending order Zn > Cr > Cu > V > Ni > Ba > Al > Sr > Cd (Table 2). The observed Pb content was lower than in the previous study (Aničić et al. 2009a) with the same species’ moss bags in the Belgrade urban area. This result coincides with the fact that leaded gasoline was banned in Serbia at the beginning of 2011. As a consequence of the introduction of unleaded fuel, Pb can no longer be taken as an indicator for current traffic-induced air pollution (Zechmeister et al. 2005). RAF values of Mg and Mn were close to 0 indicated that there was no accumulation of these elements in moss tissue; those of K and Na showed a substantial decrease after the bags exposure (Table 2), what is expected on the basis of the previous research (Aničić et al. 2009b, c).

According to the results obtained (Fig. 2), in two street canyons (MS and DJ), the vertical distribution patterns of Al, Ba, Co, Cr, Cu, Pb, V and Zn, showed statistically significant decrease from the first to the third heights of bags exposure. However, in the other street canyons (OV, KM and most elements in KN), the highest concentrations of Al, Ba, Cd, Co, Cr, Cu, Fe, Ni, Pb, Sr and V were determined in the moss exposed at the second height. Such vertical distribution could be explained by considering several important parameters that influence the pollutant dispersion process in street canyons, which include: ambient conditions (wind speed and directions), building geometry and street dimensions, thermal stratification (solar insulation and orientation, building and street thermal capacitance), vehicular movement (size, number and frequency), etc. (Xie et al. 2005). In four street canyons (DJ, KN, OV and KM), moss bags were placed on the windward sides, while in MS they were exposed on the leeward side (Fig. 3a). During the period of exposure, on the automated station (SEPA 2011), an average wind speed was hourly measured and found to be below 1.5 ms⁻¹, except in DJ Street.

There is evidence that when synoptic wind is below about 1.5 ms^{-1} , the main wind vortex within the street canyon tends to disappear and the air stagnates in the street (De Paul and Sheih 1986). In that case, a thermal vortex in the street of aspect ratio of $H/W \approx 1$ might play a dominant role whose impact depends on whether leeward or

windward side is heating. According to some researchers (Xie et al. 2005; Wang et al. 2011), when the windward side is heated, an upward buoyancy flux opposes the downward advection flux along the wall and divides the flow structure into two counter rotating cells with a clockwise top vortex and a reverse lower vortex within the canyon (Fig. 3b). It is

Fig. 3 The specific air flow in street canyons: **a** a scheme of the primary vortex and the position of exposed moss bags—4, 8 and 16 m; **b, c** the examples of thermal vortices (adapted from Wang et al. 2011) in OV and KM, DJ, respectively; **d, e** the example of dynamic vortex in asymmetric street canyon (adapted from Gu et al. 2011) in MS

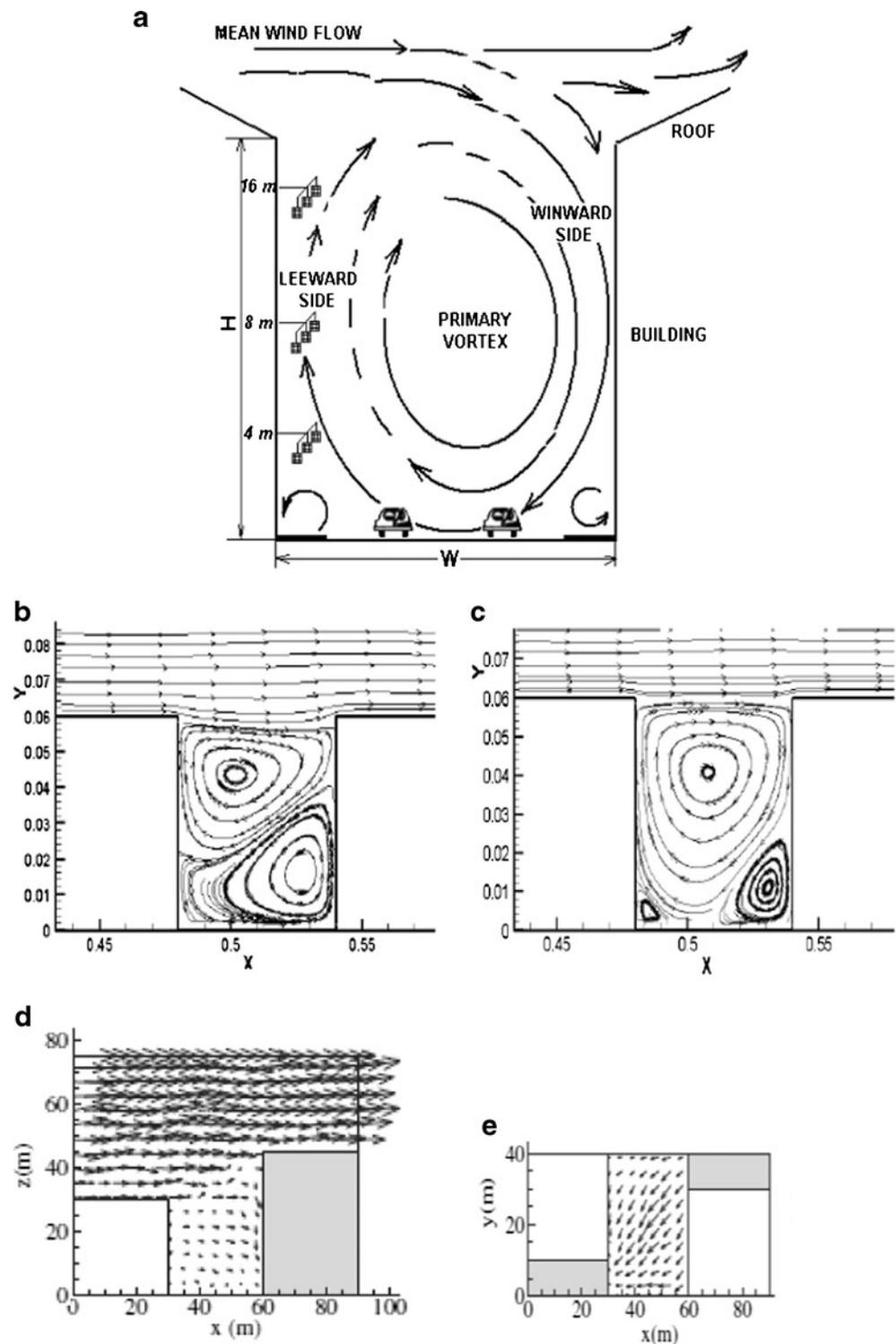


Table 3 Relative accumulation factors (RAF) of the elements in moss samples after 10 weeks of exposure in the city tunnel in Belgrade urban area

RAF	Element
≤0	K, Mn, Mg
0–1	Ca, Cd, Sr
1–5	Al, Ba, Co, Ni, Zn
5–10	Fe
10–20	Cu, Cr

most likely that, in OV and KM streets, the direction of a reverse lower vortex brings pollution from the ground and favours deposition of trace elements in vicinity of the second exposure height (at the $z/H \approx 0.29$).

On the other side, in KN Street, the highest concentration of some elements (Al, Co, Cr, Cu, Cr, Fe, Ni, Pb, V and Zn) at the second exposure height ($z/H \approx 0.29$) could be related not only to thermal vortex but also to their association with fine aerosols. Tretiach et al. (2011) found that a moss bag entraps a prevalence of potentially inhalable or breathable particles; the smallest particles classes (<2.5 and 2.5–10 μm) were largely predominant (78.8 and 19.4 %, respectively) with only 0.2 % of particles greater than 25 μm in diameter. The fine ($\text{PM}_{2.5}$) and ultra-fine ($\text{PM}_{<0.1}$) particles were reported to contain more of anthropogenic, traffic-related metals (Ni, Cr, V, Pb and Ba) than particles of other sizes, although crustal metals accounted for over 90 % of all particulate mass concentrations (Singh et al. 2002; Ntziachristos et al. 2007). Also, Lin et al. (2005) pronounced that total of 72–78 % of the Pb+Cd+Ni+Cr were retained in the fine particles and the percentage remained significant in ultra-fine ones (24–50 %). Additionally, Weber et al. (2006) demonstrated that the concentration of $\text{PM}_{2.5}$ changed significantly with height, decreasing from a maximum at $z/H=0.23$ towards both the lowest ($z/H=0.15$) and highest ($z/H=0.59$) sampling heights.

Street DJ belongs to the deep canyons ($H/W \approx 3$), in which, weak counter rotating secondary and tertiary vortices (Fig. 3c) may be observed at street level (reported by Pavageau et al. 1996; Jeong and Andrews 2002). These vortices seem to prevail over thermal vortex and may be considered as the major contributors to higher deposition of trace elements at the $z/H \approx 0.15$.

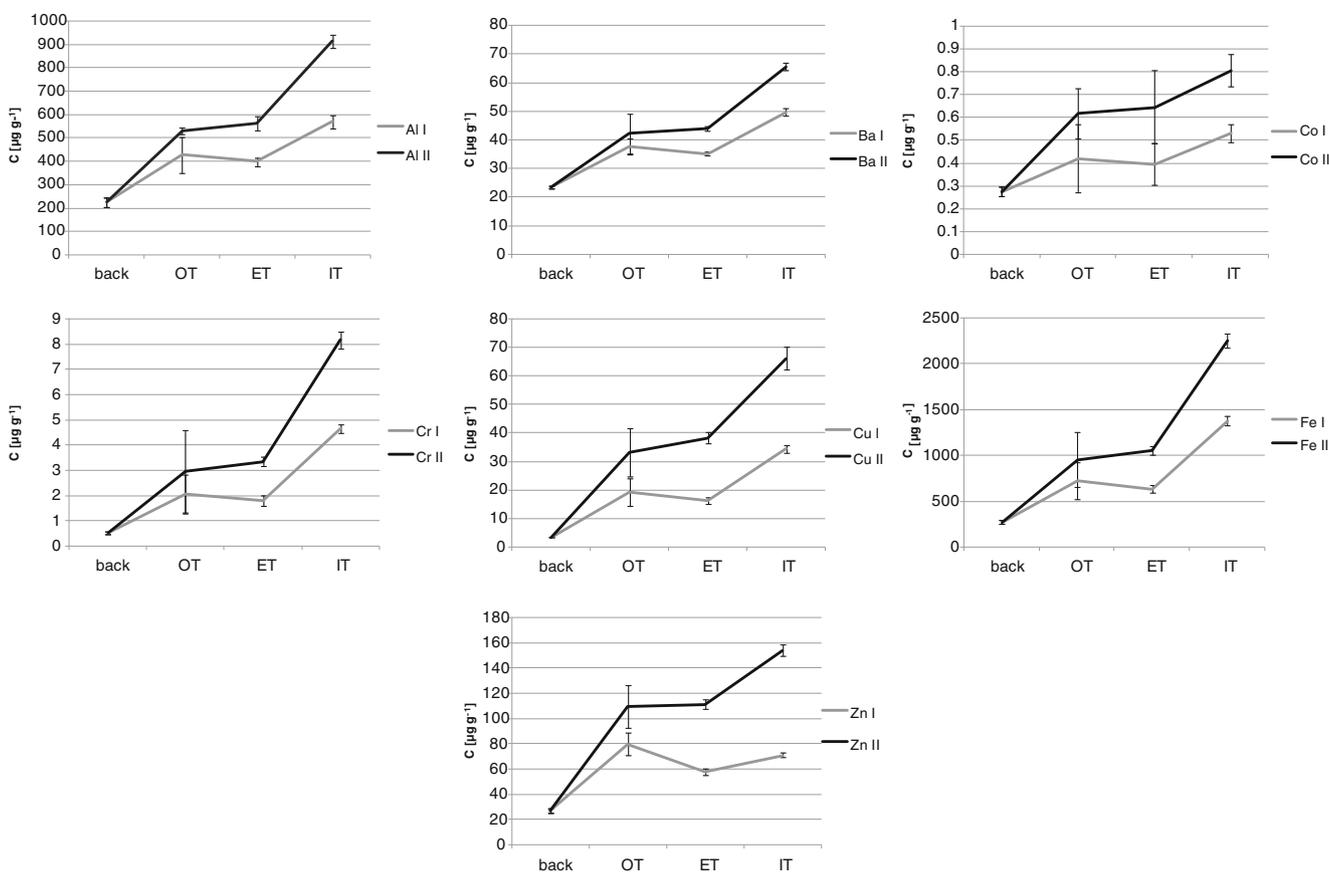


Fig. 4 Average concentrations with standard deviation bars of Al, Ba, Co, Cr, Cu, Fe and Zn in the moss bags after the exposure time of 5 and 10 weeks (I and II, respectively) in the city tunnel experiment: inside of

tunnel (IT), entrance tunnel (ET) and outside of tunnel (OT); and the initial element concentrations in the unexposed moss

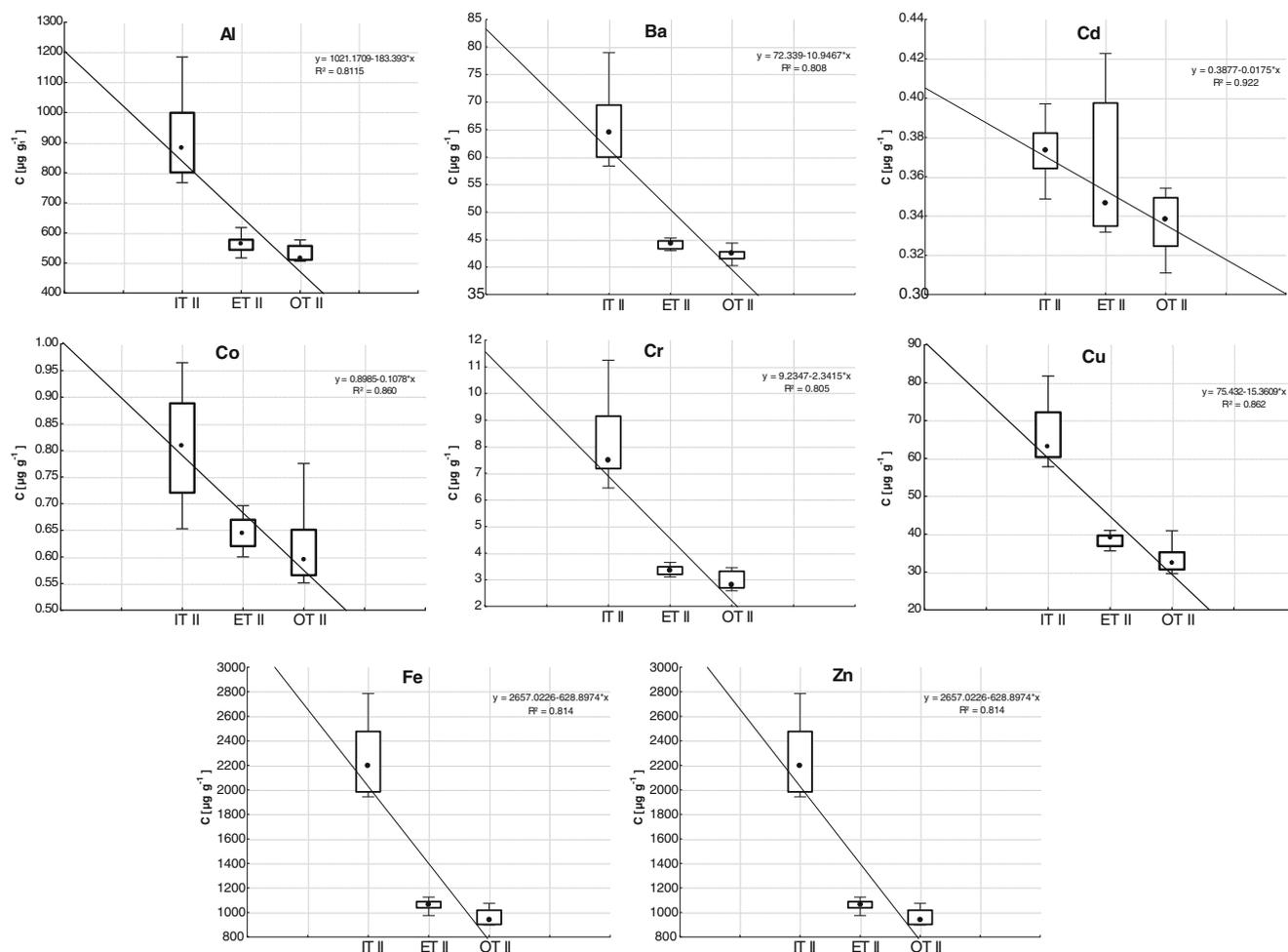


Fig. 5 Element (Al, Ba, Cd, Co, Cr, Cu, Fe, Ni and Zn) concentrations ($\mu\text{g g}^{-1}$) in the moss bags exposed for 10 weeks in the city tunnel experiment: inside of tunnel (IT), entrance tunnel (ET) and outside of tunnel (OT)

Unlike the other investigated street canyons, in MS the exposure positions were placed on the leeward side whereas the windward was heating. As a possible result of this, clockwise top vortex causes pollutant concentration to increase at the $z/H \approx 0.15$ on the leeward side. Similar to other streets, MS has $H/W \approx 1$ ratio, but there is strong asymmetry due to very high buildings on one side to those on the opposite side of a street. In such case (Fig. 3d, e), the air flow is separated at the top area of the high building wall in the step-up section, forming a strong down washing (Gu et al. 2011). Consequently, low wind speed (about 1.5 ms^{-1}) may still contribute to the highest pollution level of this street canyon.

Contrary to the often observation, the above-mentioned indicates that spatial air pollution distribution is not always uniform in microenvironments like street canyons. Obtained vertical deployment of trace elements emphasises that residents in high-level parts of some street canyons may be exposed to higher concentration of toxic and cancerogenic elements than pedestrians, cyclist, drivers, etc.

Horizontal element distribution in moss bags—tunnel experiment

In the city tunnel experiment, accumulation of trace elements in moss *S. girgensohnii* was observed after two periods of bags exposure—5 and 10 weeks. As well as in the street canyon experiment, total concentrations of Al, Ba, Co, Cr, Cu, Fe, Ni and Zn increased significantly in moss bags in comparison with their background concentrations (Table 3). Concentration of Mg and Mn were slightly increased while concentration of K and Na decreased substantially. Figure 4 shows difference in accumulation for some of above-mentioned elements between 5 and 10 weeks of exposure, which is confirmed to be statistically significant.

Depending on poor air ventilation in the city tunnel, specific horizontal element distribution in moss bags was observed. Thus, in agreement with our expectations, after 10 weeks of bags exposure, decreasing trend of concentrations in moss, exposed inside, at the entrance and outside of

the tunnel, was obtained for Al, Ba, Cd, Co, Cr, Cu, Fe and Zn (Fig. 5).

These results indicate the moss capability to provide information about pollution presence and also to reveal potentially hotspots of pollution in urban areas, such as city tunnels, necessary to the local authorities. Due to poor air conditions in the tunnel, there is a need for designing ventilation systems and implementation of regular control measurements. So, the moss bags technique has been proven to be a cost-effective method for the characterisation and monitoring of urban air contamination by some major and trace elements.

Traffic influence

Taking into account that the experimental sites are placed in the heavy traffic area, with public garages in the vicinity, traffic is expected to be a primary source of air pollution. Correlation analysis was applied to whole set of data obtained for street canyons and there was a high correlation between some elements like Al/Ba (0.76), Al/Fe (0.89), Al/V (0.80), Ba/Fe (0.88), Fe/V (0.89), Fe/Zn (0.80), Cu/Zn (0.80) and V/Zn (0.83). Therefore, most of the elements showing high correlation (Ba, Cu, Zn and V) probably indicate a major influence of traffic-related sources which have been considered already in earlier investigations (e.g. Zechmeister et al. 2005; Viana et al. 2008). On the other hand, Al and Fe are generally considered as typical geological marker elements (Sternbeck et al. 2002). However, in urban areas, they often originate from metallic wear parts of vehicles and from road dust resuspension.

In accordance to the higher traffic volume, significantly higher concentration of the elements in the exposed moss bags was pronounced for the tunnel in comparison with the street canyons. As it is expected, the obtained concentrations of majority of measured elements in KM Street (pedestrian zone) were significantly lower than in the moss hanging in the other investigated street canyons. The exceptions were the concentrations of Al, Ba and Zn which were about the concentration levels in other studied street canyons. It should be noted that the KM Street is unique due to an alley in the middle. Namely, several studies suggest that trees in street canyons reduce the air circulation and lead to higher local PM concentration (e.g. Gromke and Ruck 2007) and, consequently, lead to higher concentration of trace elements. However, among the other street canyons, the highest concentrations of elements were not found in the street with the highest traffic intensity. This contradiction leads to the conclusion that, in assessing the distribution of trace elements in street canyons, a traffic flow is not the only major factor affecting the presence and latter deployment of elements in the air. Of great importance to this process is street geometry, as well as air movement (e.g. direction of the primary vortex).

Conclusion

Significant accumulation of the elements in *S. girgensohnii* moss bags was found after 10 weeks of exposure in the urban area of Belgrade. In the street canyon experiment, a decreasing vertical element distribution from the first to the third heights of moss bags exposure was revealed. However, in two street canyons, the maximum moss element contents were obtained on the second heights. This discrepancy could be explained by both: street geometry and different direction of the thermal and dynamic induced vortices in these streets. The obtained vertical variations emphasise that residents in some streets may be more exposed to trace element air pollution than pedestrians. Thus, it is necessary to evaluate the exposure of human population in high-level apartments besides those in the breathing zones. In the city tunnel experiment, the expected decreasing horizontal (from inside to outside of the tunnel) distribution of the element concentrations in moss indicated their concentrations in the air. Also, in this experiment, the moss bags showed a positive trend of element accumulation during two periods of exposure (5 and 10 weeks) for almost all measured elements. Traffic intensity, which is considered as a major source of pollution, seems to influence a vertical trace element distribution only partially. Therefore, the spatial distribution of some major and trace elements is rather a result of the competing influence of the street geometry and, consequently, different directions of the dynamic and thermal-induced vortices and finally traffic intensity. Additionally, particle size is one of the reasons why elements derived from one source differ in their distribution.

According to the obtained results, the use of *S. girgensohnii* moss bags could be suggested as a simple, sensitive and inexpensive way to monitor the small-scale inner city spatial distribution of ambient trace element content. In addition, this study may provide valuable insights into pollution patterns of urban areas and population exposure to trace elements.

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