



Article Nature-Based Solutions to Reduce Air Pollution: A Case Study from Plovdiv, Bulgaria, Using Trees, Herbs, Mosses and Lichens

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Abstract: Nature-based solutions (NBSs) are becoming more and more recognized as useful instruments to address the challenges that urban areas are currently facing, i.e., climate change adaptation, flood mitigation, etc. In the present study, we aimed to: (1) compare the efficiency of mosses, lichens, herbs and trees in removing pollutants from the urban air using their biomonitoring potential; (2) assess their efficiency as nature-based solutions to mitigate urban air pollution; and (3) propose a framework for implementing such NBSs in urban areas. The first step involved analyses of the concentrations of 20 potentially toxic elements in eight selected biomonitors. After that, an assessment of their removal capacity was made on the basis of elements accumulation. This is the first complex study in an urban area involving the simultaneous application of organisms of eight different species and four different systematic groups (lichenized fungi, mosses, herbaceous plants, woody species) as well as such a large number of potentially hazardous elements. The present study sheds new light on some well-known biomonitors in the context of their application for air pollution mitigation. The great potential of the eight studied plant species for efficient removal of potentially toxic elements is highlighted and their implementation into NBS frameworks is recommended.

Keywords: biomonitoring; heavy metals; moss-bags; lichen-bags; ecosystem services; potentially toxic elements

1. Introduction

Urbanization is a process leading to an increased concentration of population into relatively small geographical areas, such as cities and megacities, as a result of which natural ecosystems are altered and replaced by anthropogenic ones. With the development of the city, natural habitats are fragmented, isolated and destroyed; the species composition becomes impoverished and homogenized; the hydrological regime is disturbed; and the flow of energy and the circulation of substances are modified [1,2]. Although urban areas occupy only 1–6% of the Earth's mainland, cities use a much larger share of the planet's capacity in terms of input of raw materials and release of waste. Because humanity is dependent on the biosphere for supplies of food, water, raw materials and other essential goods as well as ecosystem services, the anthropogenic-induced changes in urban areas sooner or later affect human well-being [3].

Air pollution is probably the leading environmental problem in human settlements, affecting both the population and economy, as well as the biodiversity and ecosystem processes [4]. Many chemicals, including greenhouse gases, organic compounds and fine particles, are emitted from natural and anthropogenic sources [5]. Moreover, after their release, these pollutants undergo various transformations in the atmosphere (physical, chemical, photochemical, etc.), determining their final concentrations and behavior [6]. Air pollutants do not stay close to emitters; they can diffuse long distances, depending on



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Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). the topography and meteorological conditions, transported by winds as well as vertical and horizontal temperature gradients, which turns the problem of anthropogenic air pollution from local (regional) to global. Globally, more than 5 million deaths per year are associated with air pollution [7]. A report from the WHO [8] revealed that about 80% of the urban population worldwide inhabit areas with exceedance of the hygiene norms of air pollutant concentrations.

Urban vegetation is recognized as important for cities' resilience and many other ecosystem services, but there is a lack of detailed studies and recommendations on how city planners and practitioners can take the best measures for air quality improvement [9]. Nature-based solutions (NBSs) are becoming more and more recognized as a useful instrument to address the challenges that urban areas are currently facing, i.e., climate change adaptation, flood mitigation, air purification, etc. [10,11]. The benefits of NBSs are well recognized in terms of ecosystem services assessment and include improved air quality, life quality, and human health, amongst others [12]. NBSs are especially effective in tackling environmental issues such as climate change resilience and air pollution mitigation due to their potential co-benefits [11,12]. The European Commission has defined NBSs as "Solutions that are inspired and supported by nature, which are cost-effective, simultaneously provide environmental, social and economic benefits and help build resilience" [13]. NBSs are regarded as cheap and useful instruments to address the potential hazards of climate change and environmental pollution, as they could simultaneously contribute to human health and well-being, enhance biodiversity and promote urban ecosystems resilience in the most resource efficient way [14].

Nature-based approaches (involving urban green infrastructure) could be effectively applied in reducing exposure to fine particles, potentially toxic elements (PTEs) and gaseous pollutants in urban environments. Urban vegetation is well-known to contribute to air quality monitoring [15–19] as well as to air pollution mitigation [20–22]. Some plant species with high sensitivity to environmental pollution (lichens, algae, herbs and trees) have been applied as bio-indicators of air quality for more than 40 years [23–26]. Other plant species are found to be tolerant and thus they have been used to reduce air and/or soil pollution (phytoremediation). The effectiveness of a variety of plant species for bioaccumulation of heavy metals and other toxic elements has also been assessed [4,5,15–18,27]. These authors have evaluated some species with a significant capacity for metal accumulation and tolerance to environmental pollution, thus recommending them to be used not only for biomonitoring purposes, but also for the construction of green belts, buffer green patches, green walls and others, especially in so-called "hot spot" areas (heavily polluted, mainly industrial).

The biomonitoring of air pollution, based on the analysis of micro- and macroelements in plant tissues, has been increasingly applied in Europe and worldwide [4,28–31]. One of the main advantages of biomonitors is related to their potential to act as air samplers, so called "green filters," and their widespread distribution, providing a high density of parallel measurements. The relatively easy and inexpensive sampling as well as the accumulated and time-integrated behavior of biomonitor species are prerequisites for the application of air pollution biomonitoring, especially in large-scale studies of potentially hazardous pollutants [32–34]. Phytomonitoring, as a part of biomonitoring, is a system for assessing the environmental status using the plant objects in their natural environment (passive monitoring) or after their transfer from a control to the studied area where they will be exposed for a certain period (active monitoring). Biomonitors are very often applied for the monitoring of urban pollution with potentially toxic elements as they can provide a huge number of sampling points [15,19,30,35].

The aim of the present study was to: (1) compare the efficiency of mosses, lichens, herbs and trees in removing pollutants from the urban air using their biomonitoring potential; (2) assess their efficiency as nature-based solutions to mitigate urban air pollution; and (3) propose a framework for implementing such NBSs in urban areas. Thus, the content of 20 potentially toxic elements (Al, As, B, Bi, Cd, Co, Cr, Cu, Fe, Hg, Mn, Mo, Na, Ni, Pb, Se,

Sr, U, V, and Zn) in eight biomonitors have been analyzed and their removal effectiveness have been evaluated on the basis of elements accumulation. The passive biomonitors included three deciduous trees (*Acer platanoides* L., *Aesculus hippocastanum* L. and *Betula pendula* Roth) and three herbal species (*Taraxacum officinale* (L.) Weber ex F.H.Wigg., *Plantago lanceolata* L. and *Capsella bursa-pastoris* Medik.), while the active biomonitors were one moss (*Sphagnum girgensohnii* Russow) and one lichen (*Pseudevernia furfuracea* (L.) Zopf), applied as moss-bags or lichen-bags with dried material, respectively. This is the first complex study in an urban area involving the simultaneous application of organisms of eight different species and four different systematic groups (lichenized fungi, mosses, herbaceous plants, woody species) as well as such a large number of potentially hazardous elements.

2. Materials and Methods

2.1. Study Area

The object of the present study is the city of Plovdiv, Bulgaria, which occupies the second place by area and population number in the country after the city of Sofia (the state's capital). The city of Plovdiv (42°8′9.9492″ N; 24°44′31.8048″ E) is located along the Maritsa River, in the central south part of Bulgaria. It has a population of 346,893 inhabitants (as of 2018) but the real number of citizens, including students, workers, etc. passes half a million. The city of Plovdiv is well-known as a cultural hub in Bulgaria, and this was confirmed by its recognition as the European Capital of Culture in 2019. Due to its strategic geographic position, Plovdiv is also regarded as an important industrial, transport, tourist and educational center.

The climate in Plovdiv is classified as humid subtropical, where the significant humid continental influences are manifested. The gentle winds (0 to 5 m/s) and fog that are frequently present during the cooler months are some of the factors that contribute to the deterioration of air quality and it was rated as one of the European cities with worsened living conditions since 2008 [36]. The specific topography is also a factor related to the worsened air quality (elevated PM_{10} and $PM_{2.5}$ levels) in the city of Plovdiv (barrier effect of hills, lack of winds, canyon street effect, etc.). The main inputs of atmospheric pollutants are found to be derived from traffic emissions and residential heating, while the industrial sector has a minor impact [37].

2.2. Sampling Procedure

For the purposes of the present study, the city's area was divided into urban, suburban and industrial sectors, following the urban gradient hypothesis [38], and in every one of them, two representative sites were chosen. Thus, the present study involved six experimental sites, selected according to the type of anthropogenic pressure as well as the similarity of abiotic environment (light, temperature, and exposure) and the presence of selected biomonitor species (trees and herbs). One site from the city's outskirts was selected as a control (urban background level).

Tree leaves sampling: Three trees per species per site were sampled using an Expert Tree Pruner with a telescopic handle 1.5–2.5 m (Draper Tools, Hampshire, UK) to cut off some branches from the crown. We selected branches from the outer part of the canopies and from the four sides of the tree (N, S, E, W), as well as situated 2.5–3 m above the ground. In order to prepare a representative sample, about 80 mature leaves per species per site were collected [39].

Herbs sampling: At each experimental site, the aboveground biomass of 3–4 fully grown plants per species was sampled. One composite sample per species per site was prepared for the analyses [17,18].

Moss-bags preparation, exposition and sampling: Some specimens of *Sphagnum girgensohnii* Russ. were collected from the background area of the Vitosha Nature Park ($42^{\circ}37'$ N, $23^{\circ}19'$ E, altitude 1710 m). About 3 g of unwashed plant material was placed in nylon mesh (10×10 cm) and the prepared moss-bags were exposed at the same six experimental sites for a period of 3 months [40].

Lichen-bags preparation, exposition and sampling: Lichen material (*Pseudevernia furfuracea* (L.) Zopf.) was also collected from the background area of the Vitosha Nature Park, Bulgaria ($42^{\circ}37'$ N, $23^{\circ}19'$ E, altitude 1710 m) and transported to the laboratory. The specimens were carefully cleaned, removing all bark residues. About 3 g of unwashed plant material was placed in a nylon mesh (10×10 cm) and the prepared lichen-bags were exposed at the same six experimental sites for a period of 3 months [41].

2.3. Analyses of Chemical Content

About 1 g of plant tissues were placed in 5 mL of nitric acid and left for 24 h using a microwave digestion system (Microwave Digestion System, CEM MDS 81D). The first step of wet digestion consisted of 5 min at 600 W, followed by cooling for 1 h. After that, 2 mL HNO₃ and 3 mL 30% H_2O_2 were added to the plant samples and left for another 1 h. The second step included a 10 min microwave treatment at 600 W in order to achieve full decomposition of the plant sample. The filtrate was separated carefully and double distilled water was added up to a volume of 50 mL.

The elements' concentrations were measured using the ICP-MS apparatus (Agilent 7700, DF 1000, Agilent Technologies Inc., Waldbronn, Germany). Calibration was performed by Multy VI (MERCK, Darmstadt, Germany) calibration standards. Quality control was ensured with a plant standard (NCS DC73348) [42].

The data for each site was the arithmetic mean of three samples, and the data for each individual sample was the arithmetic mean of three analytical measurements.

2.4. Bioaccumulation Patterns Analysis

To assess the bioaccumulation potential of the studied plant species, three different parameters were applied—enrichment factor, relative accumulation factor and metal accumulation factor.

2.4.1. Enrichment Factor (EF) of Plants Species Used as Passive Biomonitors

The enrichment factor (EF) values were calculated by dividing the average elements content in each plant species from the urban area by the concentration of the elements in the same species from the control area, using the formula of Mingorance et al. [43]:

$$EF = \frac{C_{urban}}{C_{control}}$$

where: C_{urban} represents the concentration of an element in the plant biomass sampled from the urban area, while $C_{control}$ represents the concentration of the same element in the plant samples from the rural/control area.

2.4.2. Relative Accumulation Factor (RAF) of Plant Species Used as Active Biomonitors

Relative accumulation factors (RAF) were applied for moss-bag and lichen-bag samples collected after 90 days of exposure at the six experimental urban sites. They were calculated by dividing the average element concentration ($C_{exposed}$) from all urban sites by the content of the same element in the unexposed plant tissues ($C_{unexposed}$), using the formula of Culicov et al. [44]:

$$RAF = \frac{C_{exposed}}{C_{unexposed}}$$

These formulas (EF and RAF) are quite similar and they correspond to the formula proposed by Ma et al. [45] for Concentration factor (CF) calculations. So, they fit well for all studied plant species and allowed us to compare their efficiency in removing potentially toxic elements from the urban air.

We used the following scale for CF to assess the environmental pollution level [46]: (1) CF \leq 1.2—no pollution; (2) 1.2 < CF \leq 2.2—low pollution; (3) 2.2 < CF \leq 3.3—medium pollution; (4) 3.3 < CF \leq 4.3—heavy pollution; (5) CF > 4.3—very heavy pollution.

2.4.3. Metal Accumulation Index (MAI)

Metal accumulation index (MAI) values were calculated according to the formula of Liu et al. [47]:

$$MAI = \left(\frac{1}{N}\right) \sum_{j=1}^{N} I_j$$

where N—total number of analyzed elements, and I_j—sub-index for variable j. The following equation was used for calculation of the sub-index:

$$I_j = \frac{x}{\delta x}$$

where x—average content of each element, and δx —standard deviation.

In the present study, we have used the MAI in order to compare the remediation ability of different plant species toward 20 potentially toxic elements from urban air.

2.5. Data Analysis

The raw data obtained have been processed using the SPSS 21 software package. The relationships between the analyzed variables have been assessed through Spearman's correlation coefficients. Student's t test was applied for discovering significant differences in PTEs accumulation by plant species as well as between the control and experimental sites (p < 0.05).

3. Results

3.1. Content of Potentially Toxic Elements (PTEs) in Plants

As a whole, all studied elements' contents varied among the plant species (Table 1), depending on their biological features. Significant differences in PTEs content were observed when comparing vascular plants (herbs) and cryptogams (moss and lichen) (*p* < 0.05). A wider range of variation was observed for Na (from 31.2 mg/kg in *B. pendula* leaves to 1539 mg/kg in *C. bursa-pastoris*), V (from 0.06 mg/kg in *A. hippocastanum* leaves to 1.58 in *S. girgensohnii*), Pb (from 2–3 mg/kg in higher plants to 28–29 mg/kg in moss and lichen), Hg (from 0.01 in *P. lanceolata* to 0.21 in *Ps. furfuracea*), B (from 5.0 mg/kg in *Ps. furfuracea* to 61 mg/kg in *A. platanoides* leaves), Zn (from 16.8 mg/kg in *A. hippocastanum* to 116.7 mg/kg in *B. pendula*), and As (from 0.25 mg/kg in higher plants to 2.18 in *S. girgensohnii*). The content of two elements was found to be less variable—Cu (from 4.53 mg/kg in *B. pendula* to 15.1 mg/kg in *Ps. furfuracea*) and Cd (from 0.23–0.24 mg/kg in tree leaves to 0.52 mg/kg in *T. officinale*).

Table 1. Average values of elements' contents in studied plant species (Plovdiv, Bulgaria).

Element	Acer platanoides		Aesculus hippocastanum		Betula pendula		Capsella bursa-pastoris		Plantago lanceolata		Taraxacum officinale		Sphagnum girgensohnii		Pseudevernia furfuracea	
	mg/kg	SD	mg/kg	SD	mg/kg	SD	mg/kg	SD	mg/kg	SD	mg/kg	SD	mg/kg	SD	mg/kg	SD
В	61.0	2.33	20.5	0.85	33.4	0.61	19.7	0.70	25.3	1.02	24.6	0.77	7.35	0.29	5.00	0.05
Na	34.9	0.96	71.2	1.25	31.2	0.41	1539	58.0	415	13.2	369.7	13.3	722	19.7	451	14.1
Al	55.3	1.21	49.0	1.23	62.7	5.69	171	6.82	224	6.28	342.3	13.5	396	11.4	405	10.6
V	0.08	0.01	0.06	0.002	0.17	0.01	0.46	0.02	0.77	0.05	0.896	0.04	1.58	0.05	1.70	0.07
Cr	0.49	0.05	0.24	0.02	0.59	0.05	1.49	0.09	1.25	0.12	2.23	0.19	1.15	0.05	1.43	0.04
Fe	128	3.71	105	3.37	130	2.69	227	11.8	340	11.8	428.9	16.8	475	8.31	529	9.12
Mn	64.0	1.72	48.7	1.56	70.6	0.68	33.0	1.21	26.6	1.04	53.3	1.69	293	5.05	100	2.06
Co	0.091	0.01	0.17	0.01	0.16	0.01	0.267	0.02	0.277	0.01	0.289	0.02	0.56	0.02	0.44	0.02
Ni	0.61	0.04	0.42	0.03	1.10	0.03	1.76	0.11	1.56	0.09	3.14	0.19	1.93	0.15	1.55	0.12
Cu	7.66	0.41	8.20	0.35	4.53	0.30	7.71	0.53	9.86	0.37	12.4	0.33	11.8	0.24	15.1	0.25
Zn	19.3	0.55	16.8	0.45	116.7	1.15	48.86	1.26	44.0	1.58	47.0	1.78	117	1.91	111	1.91
As	0.336	0.02	0.25	0.003	0.286	0.01	0.25	0.003	0.329	0.01	0.250	0.003	2.18	0.29	0.66	0.02
Se	0.486	0.03	0.44	0.03	0.300	0.003	0.30	0.003	0.600	0.04	0.357	0.01	1.58	0.15	0.30	0.003
Sr	50.4	1.46	58.6	1.59	53.43	0.38	52.29	1.43	53.0	2.14	39.286	1.36	27.9	0.54	11.5	0.24
Mo	0.229	0.01	0.28	0.006	0.33	0.01	1.46	0.06	0.786	0.1	1.029	0.04	0.88	0.21	0.20	0.002
Cd	0.231	0.01	0.24	0.012	0.243	0.01	0.434	0.02	0.306	0.03	0.516	0.03	0.49	0.02	0.37	0.02
Pb	2.71	0.07	2.75	0.075	2.32	0.02	2.214	0.09	3.529	0.15	3.143	0.12	29.3	0.48	28.0	0.53
Bi	0.31	0.01	0.29	0.003	0.82	0.03	0.216	0.01	0.271	0.01	0.200	0.002	0.89	0.09	0.20	0.002
U	0.016	0.002	0.02	0.002	0.014	0.001	0.024	0.002	0.037	0.003	0.034	0.003	0.09	0.003	0.06	0.003
Hg	0.047	0.002	0.04	0.003	0.032	0.002	0.020	0.001	0.010	0.001	0.041	0.004	0.1	0.014	0.21	0.03

3.2. Bioaccumulation Capacity of Potentially Toxic Elements by Plant

The Concentration factor (CF) values (bioaccumulation capacity) of the studied plant species in relation to the 20 PTEs are summarized in Table 2. As explained above, the bioaccumulation capacity could be assessed using different indices (EF, RAF, or CF) but all of them represent the ratio between the element's content in plants from an urban area and the content of the same element in plants from a control area.

Element	Acer platanoides	Aesculus hippocastanum	Betula pendula	Capsella bursa-pastoris	Plantago lanceolata	Taraxacum officinale	Sphagnum girgensohnii	Pseudevernia furfuracea
В	0.924	0.586	0.760	1.095	0.744	1.170	0.328	0.459
Na	2.681	2.967	0.985	9.216	4.029	9.992	1.645	6.098
Al	1.152	1.581	1.742	1.881	1.580	0.542	1.245	1.113
V	2.633	3.867	1.442	1.523	1.972	0.597	1.575	0.773
Cr	0.992	4.800	2.189	1.858	4.984	0.769	1.643	1.018
Fe	1.422	1.360	1.673	1.611	1.486	0.578	1.439	0.923
Mn	2.462	4.056	1.604	1.500	1.329	0.952	0.836	2.638
Co	1.517	0.246	0.413	0.834	1.539	0.615	1.191	1.926
Ni	0.761	1.273	0.649	1.952	1.730	1.572	1.750	1.192
Cu	0.815	1.708	1.332	1.205	0.986	0.886	2.034	1.861
Zn	0.946	1.099	0.957	1.629	1.000	0.855	1.655	1.493
As	1.344	1.000	1.144	1.000	0.411	1.000	0.702	0.332
Se	0.694	1.467	1.000	1.000	0.353	1.190	5.250	1.000
Sr	1.401	1.118	1.571	1.687	1.472	1.455	0.919	2.290
Mo	1.145	5.540	0.733	1.619	0.342	0.686	0.461	1.000
Cd	2.310	2.350	1.350	1.669	0.927	1.075	1.190	0.354
Pb	1.229	1.249	1.696	1.476	1.765	0.952	1.721	0.875
Bi	1.520	1.465	1.218	1.080	0.630	1.000	0.481	1.000
U	3.200	1.400	1.400	2.400	0.740	0.567	1.257	1.160
Hg	1.567	4.000	1.280	2.000	1.000	1.025	4.900	0.626

Table 2. Concentration factor (CF) values of studied plant species (Plovdiv, Bulgaria).

Obviously, there are some limitations to applying the assessment scale [46] when using different plant categories such as in our study. For example, the CF for Cd in *P. lanceolata, T. officinale, S. girgensohnii* and *Ps. furfuracea* has values <1.2 = no contamination; the CF for Cd in *C. bursa-pastoris* and *B. pendula* has values in the range 1.35–1.67 = slight contamination; and the CF for Cd in *A. platanoides* and *A. hippocastanum* has values between 2.2–3.2 = moderate contamination (Table 2). Nevertheless, the CF values are quite informative in assessing the potential of plant species to reduce air pollution based on their bioaccumulation capacity.

Acer platanoides was found to be the most efficient bioaccumulator of U (CF = 3.2), Bi (CF = 1.52) and As (CF = 1.34) among all studied plant species, demonstrating significant capacity (CF > 2) for Na, V, Mn and Cd, as well as moderate capacity (CF > 1.2) for Fe, Hg, Pb, Sr and Co.

Aesculus hippocastanum was proven to be an excellent bioaccumulator of Mo (CF = 5.54), Cr (CF = 4.8), Mn (CF = 4.06), Hg (CF = 4.0) and V (CF = 3.87). This species also showed a significant accumulation (CF > 2) of Na and Cd, while a moderate capacity (CF > 1.2) for Cu, Al, Fe, Ni, U, Pb, Se and Bi.

Betula pendula can be rated as the species with the lowest air pollutant removal capacity among the plants studied as no maximum accumulation was observed, as well as a CF > 2 was found for only one chemical element—Cr. However, CF values in the range 1.2–2 were obtained for 11 potentially toxic elements (Al, Fe, Mn, V, Cd, Pb, Sr, Hg, U, Bi and Cu), some of them being the highest for all trees studied—Al, Fe, Sr and Pb.

Notably, *Capsella bursa-pastoris* demonstrated CF values >1.0 for 19 of the studied 20 potentially toxic elements (except Co). The maximal value was its CF for Na (9.22), and significant (CF > 2) bioaccumulation was found for U and Hg, and it had a moderate capacity (CF > 1.2) to accumulate Al, Cr, Fe, Mn, V, Ni, Zn, Cu, Sr, Mo, Cd, and Pb.

Plantago lanceolata had a maximal capacity to accumulate Cr (CF = 4.98) and Pb (CF = 1.77) from all studied plant species, having significant potential for Na (CF = 4.03) and V (CF = 1.97), while only moderate (CF > 1.2) for Al, Fe, Co, Ni, Mn, and Sr.

In the present study, *Taraxacum officinale* was proven as the best accumulator of Na (CF = 9.99) and B (CF = 1.17) from all studied plant species, as well as a good accumulator of Ni and Sr (CF = 1.45-1.57).

The two cryptogam species also showed remarkable potential for removing PTEs from ambient air. Dried biomass of moss *Sphagnum girgensohnii* (exposed in moss-bags for a 90 day period) had the highest CF for Se (5.25), Hg (4.09), Cu (2.03) and Zn (1.66). A significant capacity (CF > 1.2) was also proven for Na, Al, V, Cr, Fe, Co, Ni, Cd, Pb and U. Dried biomass of *Pseudevernia furfuracea* (exposed in lichen-bags for a 90 day period) had the maximal CF for Mn (2.64), Sr (2.29) and Co (1.93), with excellent potential for Na (CF = 6.1). This species was found also to be a moderate (CF > 1.2) accumulator of Cu, Zn, Ni and U.

The MAI values of the eight studied plants are presented in Figure 1. The minimum was found in *P. lanceolata* (21.77), followed by *A. platanoides* (24.53); medium values were obtained for *A. hippocastanum* (33.76), *S. girgensohnii* (31.21), *T. officinale* (30.49) and *C. bursa-pastoris* (29.63); while the maximum was found for *B. pendula* (49.60) and *Ps. furfuracea* (48.47).



Figure 1. Metal accumulation index (MAI) for 20 potentially toxic elements in the studied plants.

4. Discussion

4.1. Assessment of Biomonitors' Efficiency as NBS for Air Pollution Mitigation

The MAI values obtained for Plovdiv's vegetation were quite higher when compared to some other studies [4,47,48]. Various factors could contribute to the observed differences, i.e., related to the local meteorology, local atmospheric chemistry, altitude, vegetation phase, plant performance, etc., as they affect the removal capacity of plants toward air pollutants [4,21,49]. The number of analyzed elements was also different—20 elements in our study and quite lower numbers (four to seven) of elements in the others. According to our MAI values, all studied plant species are quite tolerant of the contaminated urban environment and have significant accumulation efficiency, so they can be used as bioindicators or biomonitors for potentially toxic element contamination in urban areas. Thus, the present study confirms the other authors' recommendations [4] for the integration of such species as NBSs when constructing green buffers and barriers, for instance, between heavily impacted and vulnerable areas (hospitals, schools, kindergartens, parks, etc.).

Mosses and lichenized fungi are the most applied biomonitoring organisms worldwide. The main reason is their lack of roots, which should mean that they obtain nutrients only from the atmosphere and not from the substrate (unlike higher plants) [34,50]. However, the uptake of elements or other contributions from the substrate cannot be completely excluded [46]. Lichenized fungi and mosses are thought to accumulate elements mainly by passive processes [24,28], so the capture of relatively insoluble and metal-rich fine particles rather than the retention of soluble forms of chemical elements prevails [46,51], which is also supported by their reported high retention rates [34]. Our findings of moss-bag contents of Cu, Pb, V, and Zn correlate with the findings of Hu et al. [48] in Wuxi, China, although our Cr was significantly lower (three to four times). Our data from the lichen-bags were quite a bit higher (nine-fold for Zn, four-fold for Cu, two-fold for Pb, 1.5-fold for Cd, 1.5-fold for Mn) in comparison with the reported data from Italy [52].

Vegetation, especially herbs, are very effective biomonitors of environmental pollution (air, soil, etc.) due to their widespread distribution and availability [5,53,54]. *Taraxacum officinale* (dandelion) is one of the most commonly used species in such studies, especially in terms of pollution with potentially toxic elements, because of its large areal and ease of identification [18,55]. Concentrations of the various heavy metals in the leaves and roots of *T. officinale* collected from 132 areas in the territory of Poland were investigated by Kabata-Pendias and Dudka [56]. A high indicator efficiency of both aboveground and underground plant organs was established, different for individual elements—leaves showed higher values of Cd, Pb and Zn compared to roots. The content of Zn and Ni in *T. officinale* leaves from Plovdiv corresponds well to data from some urban localities in the USA, while the elements Cd and Cr were less accumulated [57]. When compared to some Italian cities, our results for Cr and Zn content were also lower [58].

Plantago lanceolata L. is also regarded as an excellent biomonitor due to its higher bioaccumulation potential [5,17]. Samples of roots, leaves, stalks and flowers of *P. lanceolata*, grown around a copper smelter, have been collected and the concentrations of some potentially toxic elements (Al, Cu, Fe, Mn, Ni, Pb and Zn) have been measured [5]. The data revealed that the leaves contained high concentrations of Cu, Pb and Zn (even at toxic levels), especially in plants collected from the periphery of the smelter or from the transects along the prevailing wind directions. *P. lanceolata* demonstrated a significant potential for bioaccumulation, especially in roots (Cu), aboveground plant parts (Fe) and stalks (Zn). Concentrations of some PTEs (Al, Fe, Mn, Cu and Zn) in the *Plantago lanceolata* leaves from Plovdiv were extremely higher than the results of Jordanovic et al. [5], while those of Ni and Pb were quite similar.

Aksoy et al. [59] tested the potential of *Capsella bursa-pastoris* for biomonitoring of environmental pollution. That study was carried out on the grounds of the city of Bradford, England, and the results obtained were compared with data for *Poa annua*, a species with a similar ecology, whose potential for biomonitoring has been explored in previous studies. The authors confirmed the qualities of both species based on the high linear regression for Pb, Cd, Zn and Cu contents in soil and leaves (after washing), as well as a consequence of the possibility to distinguish atmospheric deposition from root exchange.

Tree leaves are well-known as reliable biomonitors to track air pollution with trace elements [4,15,16,27,35,60], although there are difficulties that arise when comparing data from different studies. The problems are connected to both the use of various tree species and various experimental methodologies [19]. Leaf accumulation of PTEs in some broadleaf species (*Acer platanoides, Aesculus hippocastanum, Betula pendula*, and *Tilia cordata*) from high-traffic areas in Belgrade, Serbia, was studied by Tomašević et al. [19]. The Serbian leaf contents of Cr, Cu, Fe and Pb were significantly higher than values found in the same plant species from Plovdiv (except Zn in *B. pendula*). Our data for Cu and Cd leaf contents in *A. hipocastanum* and *B. pendula* correspond well to the results from Wroclaw, Poland [61], although the Pb content was two and three-fold higher (in *B. pendula* and *A. hipocastanum*, respectively). *A. hipocastanum* leaves, collected from urban, suburban and roadsides in the Thrace region of Turkey (Istanbul, Edirne, Tekirdag, Corlu), have been studied by

Yilmaz et al. [62] and the concentrations of Pb, Cd, Zn and Cu are similar to our findings. Another study from Turkey [63] also evaluated the leaf contents of Cd, Ni, Pb and Zn (*A. platanoides, A. hippocastanum* and *B. pendula*), but the samples had been collected from the city of Istanbul.

According to Kabata-Pendias and Pendias [64], the Pb content of tree leaves (both deciduous and coniferous) in Europe varied in the range of 1.5–2.1 mg/kg. The highest Pb value, found in our study, was 5.0 ± 0.1 mg/kg (urban zone), which means that the urban air is quite polluted with this element (based on foliar bioaccumulation) in comparison with other European cities (2.5-fold higher).

4.2. Framework for Implementing NBSs to Mitigate Air Pollution in Urban Areas

Here we provide some recommendations to help planners and practitioners to apply the best measures when implementing NBSs for air quality improvement as well as for enhancing the ecosystem services derived by urban green infrastructure.

The first step in the proposed framework for NBS implementation (Figure 2) requires the identification of the priority urban areas with more deteriorated air quality. The next step is to elaborate the major sources of air pollution, urban meteorology, microclimate and heterogeneity. The third step includes the selection of a NBS strategy, which mainly depends on the location characteristics (topography, microclimate, etc.), type of source (domestic heating, traffic or industrial pollution) and the desired outcome (air purification, noise attenuation, urban heat island mitigation, etc.). After that, in step 4, a careful selection of plant species based on their adaptation and tolerance to an urban environment, physiological characteristics, and air pollutants removal efficiency will be crucial in the mitigation of air pollution to a greater extent [16–18,22,28].



Figure 2. Framework for NBS implementation in urban areas for air pollution mitigation.

There is no doubt that plants and notably trees are particularly efficient solutions in removing pollutants from both air and soil [20,22,65]. They have large crowns, with a huge leaf area, and they act as pollutant capture devices for both gaseous and particulate substances from the ambient air. Hence, urban trees are found to be one of the most suitable NBSs for reducing roadside air pollution by both leaf surface retention, stoma absorption and wax accumulation [66]. Field measurements have shown that a wide vegetation protection belt along a road can markedly reduce the fine particle (PM₁₀ and

 $PM_{2.5}$) concentration. Avenue trees have lowered the PM_{10} levels by 39–89% (depending on the wind rose) and the $PM_{2.5}$ level by 17% [66]. Other benefits are related to the wind speed retention, noise attenuation, shading of sidewalks and bicycle lanes, lessened health risk, etc. Accordingly, the planting of roadside vegetation should be included in all NBS strategies for achieving urban sustainability. A medium spacing (approximately equal to the crown diameter) offers the highest impact in reducing air pollution on both bicycle lanes and roads [66]. Evergreen species have an advantage in filtering the air all year long [4,22], but deciduous trees have wider crowns and a larger leaf area, thus greater trapping efficiency [67]. Some previous studies also showed that urban vegetation is permanently exposed to the presence of plenty of toxic gases, aerosols and particulates, which even at chronic concentrations seriously affect plant physiology. For this reason, plants are involved in a constant process of adaptation toward the dynamics of an urban environment, aiming to achieve optimal development. There is a lack of knowledge about the proper adaptation mechanisms that take place, but some different signaling pathways should play role. In our previous study, we have proposed a reference scale of biochemical markers for the assessment of plant stress induced by urban air pollution [68]. Furthermore, they can be applied in landscaping activities and strategies for the sustainable management of urban forestry.

Various herb species are used for the construction of urban lawns, green walls and green roofs, especially when some limitations to tree development or maintenance problems persist [13]. Due to their biology, herbs are preferred over trees for soil phytoremediation purposes [10,69–71]. Urban lawn construction is a typical example of a NBS, aiming at sustainable management of urban soils, providing low-cost remediation, biodiversity support and other benefits. Herbal cover (i.e., urban lawns) could improve the soil structure by enhancing its porosity and aggregate stability, as well as by increasing the organic matter content and water holding capacity. It also influences many soil processes, soil functions, and soil microbiota as well, thus contributing to a more resilient soil ecosystem that has a better buffer capacity against external impacts and creates better livelihood conditions. Our previous work [72] describes an algorithm for urban lawns and buffer green patches construction. These buffer patches along roads led to a significant decrease in the trafficderived pollutants that have been deposited on soil surface via ambient air. Furthermore, they exerted a positive effect on the soil microbiota's structure, enzymatic activities, etc. Furthermore, the sustainable management of urban soils is related to that of the urban air due to the carbon sequestration, greenhouse gases reduction, dust suspension and retention, and thus benefits air quality and human health. So, NBS integration into urban planning and management can lead to a significant enhancement of plenty of ecosystem services in urban areas.

In recent years, mosses are preferred and widely used for green walls construction in both indoor and outdoor conditions [71]. However, there are a lot of problems related to their indoor survival and their horticulture [73]. At the same time, their harvesting from nature is crucial to the environment and habitats [73]. In such cases, the usage of dried moss and/or lichen material can be implemented as an alternative to the living material without any need for periodic replacement. Furthermore, these non-vascular plants can accumulate plenty of PTEs from urban air in their dried tissues, thus reducing air pollution, especially in so-called hot spots. Possible removal of accumulated pollutants by washing can be applied to achieve recycling and reuse of dried wall material. That can also be an option for places with very poor environments where no plant will survive.

5. Conclusions

Urban vegetation is one of the most effective NBSs for reducing urban pollution and it provides many other ecosystem services to human well-being. A careful selection of plant species and design parameters, as well as proper planning of NBS strategies according to local conditions is needed for their successful and effective performance. The synergy between anthropogenic and natural processes when implementing NBSs reveals lower costs and lower maintenance, and they may even be more effective in achieving urban resilience.

- (1) The present study sheds new light on some well-known biomonitors (trees, herbs, mosses and lichens) in the context of their application for air pollution mitigation.
- (2) Ornamental trees demonstrated significant bioaccumulation of Al, Fe, and Pb (*Be-tula pendula*); Mo, V, and Cr (*Aesculus hippocastanum*); and As, Bi, and U (*Acer pla-tanoides*), having Metal accumulation index (MAI) values in the range of 49.6, 33.76, and 24.53, respectively.
- (3) Herbal species were found to be excellent accumulators of Na and B (*Taraxacum officinale*); Na, U and Hg (*Capsella bursa-pastoris*); and Cr and Pb (*Plantago lanceolata*). Their MAI values were in the range of 30.49, 29.63 and 21.77, respectively.
- (4) Moss (*Sphagnum girgensohnii*) and lichen (*Pseudevernia furfuracea*) dried materials were the best accumulators of Se, Hg, Zn, and Cu (moss) and Mn, Sr, and Co (lichen), with MAI values of 31.21 and 48.47, respectively.

The great potential of the eight studied plant species for efficient removal of potentially toxic elements is highlighted and their implementation into NBS frameworks is recommended.

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