Article

Accumulation Characteristics of Metals and Metalloids in Plants Collected from Ny-Ålesund, Arctic

Hongmei Ma [1], Guitao Shi [2],* and Yongqian Cheng [3]

[1] Ministry of Natural Resources Key Laboratory for Polar Science, Polar Research Institute of China, Shanghai 200136, China; mahongmei@pric.org.cn

[2] Key Laboratory of Geographic Information Science (Ministry of Education), School of Geographic Sciences and State Key Lab of Estuarine and Coastal Research, East China Normal University, Shanghai 200241, China

[3] Chinese Research Academy of Environmental Sciences, Beijing 100012, China; chyongqian@163.com

* Correspondence: gtshi@geo.ecnu.edu.cn; Tel.: +86-21-54341208

Abstract: Toxic elements can be transported to polar regions by long-range atmospheric transport from mid and low latitudes, leading to enrichment of elements in the polar environment, especially in the Arctic. The plants can be ideal bioindicators of element contamination in environments, but information on the element enrichment and sources of plants remains limited in polar regions. Here, concentrations of 15 metals and metalloids (Pb, Ni, Cr, Cu, Co, As, Cd, Sb, Hg, Se, Fe, Zn, Mn, Al, and Ti) in six species of plants, *Deschampsia caespitosa* (Tufted Hair Grass), *Puccinellia phryganodes* (Creeping Alkaligrass), *Saxifraga aizoides* (Yellow Mountain Saxifrage), *Dicranum angustum* (Dicranum Moss), *Salix Polaris* (Polar Willow), and *Cerastium arcticum* (Arctic Mouse-Ear Chickweed), collected from Ny-Ålesund, the Arctic, were determined, and enrichment and sources of elements were assessed. Results show that element concentrations vary in different plant species, and element levels in *D. angustum* and *C. arcticum* are generally higher. In spatial terms, elevated element concentrations were found near residential areas, while low element levels were present at the sites far from settlement points. Enrichment assessment shows that Cd, Hg, and Zn are the most enriched elements, with enrichment factors above 30, suggesting sources other than soil dust control their concentrations. Principal component analysis (PCA) showed that the extracted three components can explain 82% of the total variance in element concentrations. The elements Ni, Cr, As, Sb, Fe, Al, Ti, and to a lesser extent Co are highly loaded in PC1, possibly associated with continental crust particles. PC2 is closely correlated with Cd, Se, Mn, Cu, and Zn, while Hg and Pb have high loadings on PC3. The elements highly loaded on PC2 and PC3 are likely associated with pollutants from atmospheric transportation. Together with enrichment assessment, the investigated plants have a great potential for monitoring atmospheric Cd, Hg, and Zn pollution in Ny-Ålesund, and *D. angustum* and *D. caespitosa* are the more sensitive species. The results would be of significance for monitoring element contamination in the pristine Arctic environments using the bioindicator plants.

Keywords: element; plant; enrichment factor; Arctic

1. Introduction

The pristine Arctic has become a region of great concern, and growing evidence has indicated that anthropogenic contaminants (e.g., heavy metals) can reach the remote Arctic from mid and low latitudes via long-distance atmospheric transport [1–6]. In the Arctic, increased contaminant concentrations have

been found in varied environmental compartments, such as atmosphere, snow, soil, and sediment [7–12]. Among the contaminants, mercury (Hg) has been attracting more attention, considering that Hg, once deposited, can be methylated by bacterial activities and becomes bioavailable to organisms, subsequently potentially affecting the ecosystems [13,14]. Similar to Hg, other potentially toxic metals, such as lead (Pb) and cadmium (Cd), can also biomagnify via the food chain, and accumulate in living organisms [15]. Therefore, the investigations on occurrence, concentrations, and distribution patterns of toxic metals in the Arctic ecosystem are critical to evaluate to what extent human emissions influence this pristine environment.

Plants, especially mosses, with weak or absent cuticle and very thin leaves, favoring the exchange between atmosphere and cell walls, are cheap and sensitive metal and metalloid biomonitors. In previous studies, the bioaccumulation of metals was extensively investigated, e.g., [16,17]. Among the biological species used for metal pollution biomonitoring, mosses have the most common occurrence. Due to a number of advantages, such as no seasonal variation of the morphology and considerable longevity [18], mosses could be the best long-term integrators of the atmospheric deposition of elements. During recent decades, the biomonitoring role using moss as indicators of metal pollution has been widely reported [19–22]. For instance, in Europe and North America, mosses have been extensively used for monitoring atmospherically transported heavy metal deposition [23–26]. So far, most of the investigations have mainly been focused on element concentrations near human settlements; the investigations on metal levels in plants in the remote polar regions, however, remain limited.

Svalbard, a Norwegian archipelago in the Arctic Ocean, is one of the most northern places where humans live. The atmospheric contaminants emitted from local human activities are thought to be minor. However, pollutants from the industrialized regions can be an important factor concerning Svalbard, due to the long-distance atmospheric transport. Up to now, only a few investigations have been performed to characterize metal accumulation in soils and mosses in Svalbard. For instance, the report on metals in mosses and lichens in southwestern Svalbard suggested that long-distance transport of anthropogenic emissions was responsible for metal accumulation [27]. Ny-Ålesund, located in the northern part of Svalbard, is the northernmost functional civilian settlement in the world. Jia et al. (2012) have examined antimony (Sb) concentration in topsoils and mosses in Ny-Ålesund [28]. The information on element accumulation and sources in plants (mosses and angiosperms), however, remains limited. Here, we carried out an investigation of a comprehensive set of elements in six species of plants in Ny-Ålesund, Svalbard. Concentrations of 15 elements, Hg, Pb, Cd, chromium (Cr), copper (Cu), arsenic (As), antimony (Sb), selenium (Se), nickel (Ni), zinc (Zn), cobalt (Co), ferrum (Fe), manganese (Mn), aluminum (Al), and titanium (Ti) were determined, and their enrichment and main sources in plants were evaluated. Accordingly, the plant species that would be ideal bioindicators of metals in the Arctic were distinguished. The results would be of help for a better understanding of element accumulation in different environmental compartments in the Arctic ecosystem.

2. Experiments

2.1. Study Area

The Svalbard Archipelago (74°–81° N, 10°–35° E) is located on the boundary between the Norwegian Sea, the Barents Sea and the Arctic Ocean. Previous studies showed that Svalbard is perhaps one of the regions most affected by anthropogenic pollution transported from industrialized areas [2]. Ny-Ålesund, located on the west coast of Spitsbergen, is a former Norwegian coal mining town and was closed in the 1960s due to a tragic accident [29]. At present, there are about several tens of all-year permanent residents (scientific expeditioners and logistical support personnel) in Ny-Ålesund, and the population reaches more than one hundred in summer. The general energy of home heating is from diesel generators, which can emit pollutants and consequently influence the local environments potentially. Ny-Ålesund is warmer and more humid than elsewhere with similar latitude in the Northern hemisphere due to the changes of atmospheric circulation and oceanic cycle in the North

Atlantic and the Barents Sea. Annual mean temperature in Ny-Ålesund is approximately 4 °C, and most Arctic animals and plants can be found there [30]. The wind at Ny-Ålesund is predominantly from south-westerly directions [31]. Because of the unique climate qualities and geographical location, Ny-Ålesund is thought to be an ideal location for Arctic scientific exploration. Thus far, a number of research stations have been established in Ny-Ålesund; amongst them is the Chinese Yellow River Station (78°55′ N, 11°56′ E), which was built in 2004 (Figure 1).
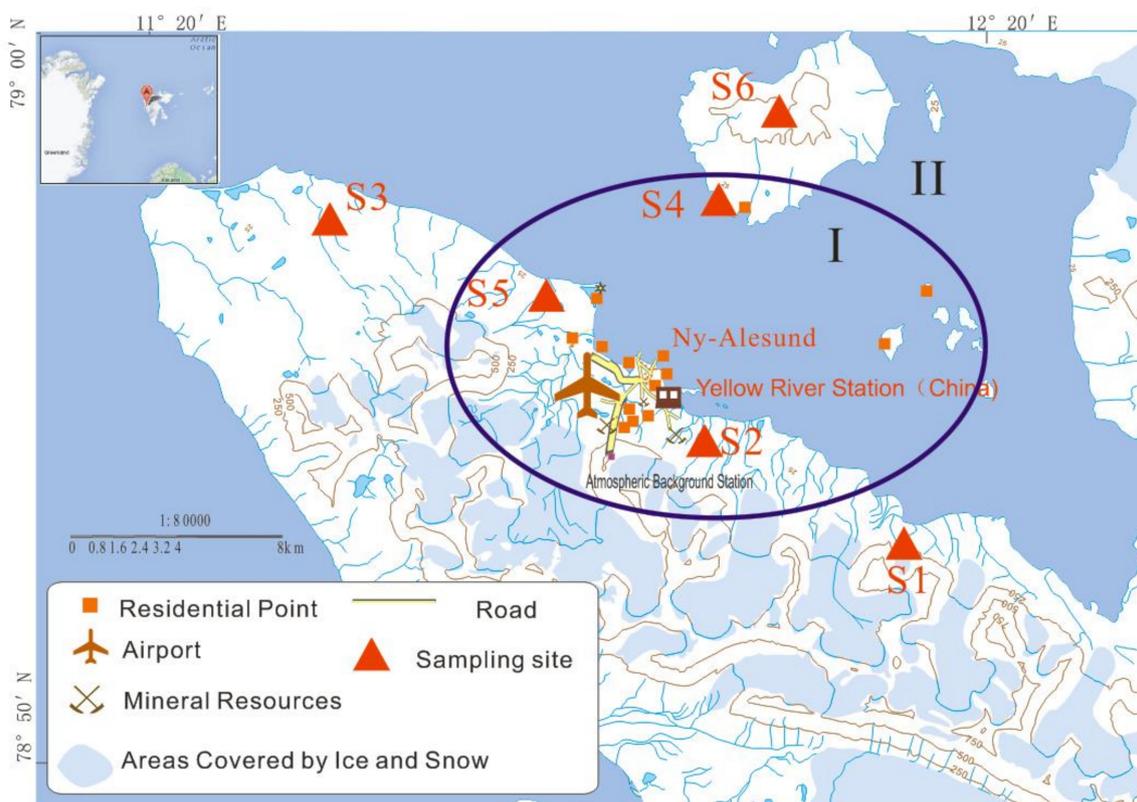

Figure 1. Sketch map showing the sampling locations in Ny-Ålesund, the Arctic. The six sampling sites (closed triangles) were classified into two groups (I and II), i.e., the sampling sites close to the residential points (closed squares), namely S2, S4, and S5, were classified into Zone I, while the stations further away from the residential area are in Zone II. The mineral resources refer to the coal mines. The contour lines were also shown as solid brown lines, with altitudes marked. The inserted panel at the upper-left corner represent the relative location of the study area in the Arctic.

2.2. Sample Collection

In Ny-Ålesund, six sampling stations, i.e., S1–S6, were selected for sample collection (Figure 1). At each investigation station, different species of plants were collected around an area of ca. 100×100 m. In the study area, in total six plant species were found and sampled, i.e., *Deschampsia caespitosa* (Tufted Hair Grass; at S1, and S2), *Puccinellia phryganodes* (Creeping Alkaligrass; at S1, S3, and S4), *Dicranum angustum* (Dicranum Moss; at S1–S6), *Saxifraga aizoides* (Yellow Mountain Saxifrage; at S2 and S4–S6), *Salix Polaris* (Polar Willow; at S3), and *Cerastium arcticum* (Arctic Mouse-Ear Chickweed; at S3, S5, and S6). Among the six plant species, *Dicranum angustum* is moss while the other species are angiosperms. It is noted that only plant leaves have been collected because these leaves are most sensitive to air pollutants. At each sampling station, leaves of the same species were collected and combined to represent one sample, with a weight of ~200 g. The sampling time was September, and the plants were mainly at end growth stage. In total, 19 plant samples were collected. The plants were kept in polyethylene zipper bags to avoid contamination, and then they were stored at −20 °C and

transported to the laboratory for treatment and analysis. While the number of sampling sites is rather limited, i.e., only six investigation stations, due to the limited logistical support (Figure 1), this study provides an opportunity to characterize the general element accumulation in plants in the remote Arctic ecosystem.

2.3. Sample Analysis

Considering that the main aims of the study are to characterize the bioconcentration of elements in plants and their potential for indicating atmospheric contamination as bioindicators, the plants were cleaned and washed with Milli-Q water (18.2 MΩ) 5 times in laboratory [32–34]. Then the plants were freeze-dried at −20 °C (ALPHA 1–4/LD, Martin Christ Inc, Osterode am Harz, Germany), until a constant weight was achieved. All the dried samples were mixed thoroughly with the aid of agate mortars and pestles. About 0.5 g of the samples were picked and placed in PTFE vessels. Plants were digested with HNO_3 and H_2O_2 (*v*/*v* = 4), and a microwave oven (Ultra WAVE, Milestone Inc, Bergamo, Italy) was used for the digestion following operations of 30 °C (5 min), 150 °C (5 min), and 200 °C (20 min). Then the solutions were dissolved with 1% HNO_3 into a final volume of 50 mL for element concentration determination.

Concentrations of Pb, Ni, Cr, Cu, Co, As, Cd, Sb, Hg, Se, Fe, Zn, Mn, Al, and Ti were determined using inductively coupled plasma mass spectrometry (ICP-MS) (PE-Sciex DRC II, Perkin Elmer Inc, Waltham, USA). In general, the instrument analyzing conditions are similar to those in Shi et al. (2015). Briefly, the solutions were introduced to the plasma using a peristaltic pump by a nebulizer and spray chamber. The operating parameters for ICP-MS were RF-1100W, the auxiliary gas flow rate 1.80 L min^{-1}, the nebulizer gas flow rate 0.95 L min^{-1}, the plasma gas flow rate 15 L min^{-1}, the scan mode was peak hopping, and the lens voltage 6.5 to 9.0 V. The elements As and Se were determined using a reaction–collision cell, with He and H_2 gases for As and Se measurement, respectively. In addition, Rhodium was used as the internal standard to examine the instrument responses.

All the analyses were run under an analytical quality protocol, including the analysis of procedural blanks, duplicate samples, and certified reference materials (the biologic reference material GBS10023, provided by the National Research Center for Certified Reference Materials, China). The recovery of the 15 elements in the reference materials varied from 89% to 105%. However, replicate analyses of plants (*n* = 3) showed high standard deviations (up to 16%), which is possible because the plants are not so easy to clean mechanically concerning the dust particles. This is consistent with previous investigations on element concentrations in lichens [35]. The detection limit (DL) was defined as 3 standard deviations of replicate runs of 1% HNO_3 (v/v) (*n* = 11). The values of the DL were 0.003, 0.003, 0.002, 0.023, 0.001, 0.014, 0.002, 0.0002, 0.001, 0.042, 0.03, 0.044, 0.013, 0.091, and 0.037ng mL^{-1} for Pb, Ni, Cr, Cu, Co, As, Cd, Sb, Hg, Se, Fe, Zn, Mn, Al, and Ti, respectively, which correspond to the element contents of 0.3, 0.3, 0.2, 2.3, 0.1, 1.4, 0.2, 0.02, 0.1, 4.2, 3.0, 4.4, 1.3, 9.1, and 3.7 µg kg^{-1} in the samples, respectively. In this study, the element concentrations in plants were above the DL. In general, the procedural blanks, which were prepared using the same protocols as those of samples but without adding sample, were lower than or close to the DL.

In addition, all the containers used in the analytical procedure were washed with dilute HNO_3 (20%, *v*/*v*), followed by Milli-Q water, in order that they are free of contamination, and all personnel wore disposable PE gloves to minimize the potential contamination during sample treatment and analysis.

2.4. Enrichment Factor Analysis

In order to remove fluctuations of absolute values and to give a clue to the sources of elements, raw concentrations were sometimes normalized to the soil or the Earth's crustal abundance pattern by calculating the enrichment factor [18,36]. Mathematical expression of the enrichment factor (EF) is as follows:

$$\text{EF} = \frac{C_n(\text{plant})/C_{ref}(\text{plant})}{R_n(\text{reference})/R_{ref}(\text{reference})} \tag{1}$$

where C_n(plant) and R_n(reference) are concentrations of elements in plant and reference environment, respectively; C_{ref}(plant) and R_{ref}(reference) are concentrations of the reference element in plant and reference environment, respectively. Enrichment factor (EF) can be classified into seven levels, i.e., (1) EF ≤ 1.5 (no enrichment), (2) 1.5 ≤ EF ≤ 3 (minor enrichment), (3) 3 ≤ EF ≤5 (moderate enrichment), (4) 5 ≤ EF ≤ 10 (moderately severe enrichment), (5) 10 ≤ EF ≤ 25 (severe enrichment), (6) 25 ≤ EF ≤ 50 (very severe enrichment), and (7) EF > 50 (extremely severe enrichment). Elements with low occurrence variability, such as Ti, Al, Fe, and Ca, were often chosen as the reference element for EF calculation [37–39]. In this study, metal Al was selected as the reference element for its high concentration and low spatial variation in the study area. For evaluating the enrichment of elements in plants, the local soils are a better reference environment. The soils in Ny-Ålesund, however, were not sampled. Here, the Spitsbergen topsoil was selected as the reference environment, and the element concentrations are from the published data [27,28]. Elements for which soil is the only source have EFs of ~1.0, while elements from human emissions are expected to have EFs > 1.0. Because the types of soils vary spatially and the possible fractionation of elements occurs during weathering, only elements with EFs < 10.0 are considered as having a crustal source [40,41].

2.5. Multivariate Statistical Analysis

Multivariate statistical analyses, such as principal component analysis (PCA) and correlation analysis (CA), are useful tools in element source apportionment. Multivariate statistical analyses of the data in this work were carried out by means of SPSS v.11.5 software packages. The essence of PCA is converting the observed variables into factors or principal components, so that a minimized set of underlying variables can be identified. Bartlett sphericity test ($p < 0.001$) indicated that the data were suitable for PCA. The raw element content data, in general, do not follow a normal distribution ($p > 0.05$, one-sample Kolmogorov–Smirnov test), but they fit the logarithmic normal distribution. Thus, the raw content data were normalized by logarithmic transformation for the PCA. Varimax with Kaiser normalization rotation was applied to maximize the variances of the factor loadings across variances for each factor, and the regression method was selected for calculating the factor score coefficient. During the PCA, the components with eigenvalue >1.0 were extracted. The loadings were calculated from the eigenvalues of the components and their corresponding eigenvectors. For a test of the stability of PCA results, one sample was arbitrarily removed from the full samples, and the PCA outcome was close to that for the full cases.

Correlations between the original variables were present in the form of non-parametric Spearman correlation coefficients.

3. Results

Element concentrations in plants are summarized in Table 1. As for the means, element concentrations in plants vary in the decreasing order of Al > Fe > Ti > Mn > Zn > Cu > Pb > Ni > Cr > Co > As > Cd > Se > Sb > Hg, which does not follow the same patterns as decreasing upper continental crustal abundance [42], possibly suggesting non-crustal sources (i.e., human inputs) and/or the varied bioavailability of elements. The variation coefficients of elements (Cv, one standard deviation versus mean) in plants are all above 0.5 (ranging from 0.5 to 1.3), implying moderate-high variation.

Element concentrations in plants from other areas in Arctic and other circumpolar Arctic regions are summarized in Table 2. In terms of continental crust-derived elements (e.g., major elements Al, Fe, and Ti), a spatial variability was present in different study areas, e.g., Fe concentration in the plants of Sweden (117 mg kg^{-1}) was much lower than that of Taimyr Peninsula (2640 mg kg^{-1}). In general, concentrations of the major elements in this study are comparable to the values of Taimyr Peninsula and higher than the other reports. Concentrations of Pb, Ni, Cr, Cu, and Cd in this study fall within the published ranges, while concentrations of As, Sb, Hg, and Zn are generally higher than the values for other regions. It is noted that element concentrations in this study are comparable to those in SW Spitsbergen.

Table 1. Statistics of element concentrations in plants ($n = 19$)/mg kg^{-1}.

Element	Mean	Minimum	Maximum	Standard Deviation	Coefficient of Variation
Pb	3.70	0.50	17.90	4.70	1.27
Ni	2.90	0.50	7.40	2.10	0.72
Cr	2.10	0.40	8.50	2.30	1.10
Cu	5.00	1.20	12.00	2.70	0.54
Co	0.99	0.12	4.50	1.11	1.12
As	0.40	0.06	1.46	0.40	1.00
Cd	0.20	0.04	0.77	0.21	1.05
Sb	0.13	0.03	0.26	0.08	0.62
Hg	0.06	0.01	0.16	0.05	0.83
Se	0.20	0.01	1.34	0.31	1.55
Fe	1264	181	4445	1250	0.99
Zn	44	17	114	29	0.66
Mn	154	27	494	141	0.92
Al	2690	352	8686	2101	0.78
Ti	166	17	644	163	0.98

Table 2. Comparison of element concentration means in plants of this study and other reports/mg kg^{-1}.

Region	Pb	Ni	Cr	Cu	As	Cd	Sb	Hg	Fe	Zn	Mn	Al	Ti	Reference
Ny-Ålesund	3.7	2.9	2.1	5.0	0.40	0.20	0.13	0.06	1264	44	154	2690	166	This study
NW Canada	6.75	3.19	3.88	3.54	0.37	0.19	0.029	n.a.	n.a.	21.24	n.a.	n.a.	n.a.	[43]
Canadian High Arctic	2.0	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	[44]
Southern Spitsbergen	7.07	4.25	n.a.	6.01	n.a.	0.59	n.a.	n.a.	n.a.	21.13	n.a.	n.a.	n.a.	[45]
Taimyr Peninsula, Siberian Arctic	1.84	4.24	n.a.	4.57	0.39	0.166	n.a.	0.051	2640	27.5	113	2820	220	[8]
Finland, 2005	2.70	1.45	0.91	3.08	0.11	0.14	n.a.	0.040	186	31.6	n.a.	176	n.a.	[25]
Iceland, 2005	1.35	3.15	3.33	7.70	0.11	0.052	n.a.	n.a.	n.a.	21.2	n.a.	n.a.	n.a.	[25]
Norway, 2005	2.17	1.24	0.58	4.37	0.12	0.089	0.070	0.054	273	31.4	n.a.	255	n.a.	[25]
Russian Federation, 2005	n.a.	2.74	3.64	8.94	0.23	0.24	0.12	n.a.	679	40.1	n.a.	850	n.a.	[25]
Sweden, 2005	2.15	0.61	0.61	3.56	0.065	0.14	n.a.	n.a.	117	30.6	n.a.	n.a.	n.a.	[25]
Ny-Ålesund	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	0.108	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	[28]

(1) n.a., not available.

It is thought that element contents can vary among plant species at the same site, possibly due to the different structures of plants [18]. In this case, interspecies comparison is essential for monitoring element pollution with the aid of plants. Element concentrations in the six species of plants are shown in Figure 2, and the values vary significantly among different species, with the maximum/minimum ratios of 9.7 (Pb), 3.9 (Ni), 8.9 (Cr), 3.7 (Cu), 6.0 (Co), 7.6 (As), 4.5 (Cd), 7.4 (Sb), 6.1 (Hg), 25.8 (Se), 8.9 (Fe), 3.0 (Zn), 7.4 (Mn), 5.4 (Al), and 8.6 (Ti). In general, elevated concentrations were found in *D. angustum* and *C. arcticum*, with the highest means of Pb, Ni, Co, As, Cd, Se, Fe, and Zn in *D. angustum* and the highest means of Cr, Sb, Hg, Al, and Ti in *C. arcticum* (Figure 2). Thus, these two species of plants, especially the moss *D. angustum*, which was widely distributed in Ny-Ålesund (i.e., sampled at all of the six investigation stations), could be a more effective biomonitor of atmospheric deposition of elements. On the other hand, element contents are generally higher in the moss *D. angustum* than in other species (angiosperms), also suggesting a higher degree of element enrichment in moss than in angiosperms. It is noted that element concentrations in *S. Polaris* could be unrepresentative of the general concentration patterns in Ny-Ålesund, considering that only one sample was included in the interspecies comparison.
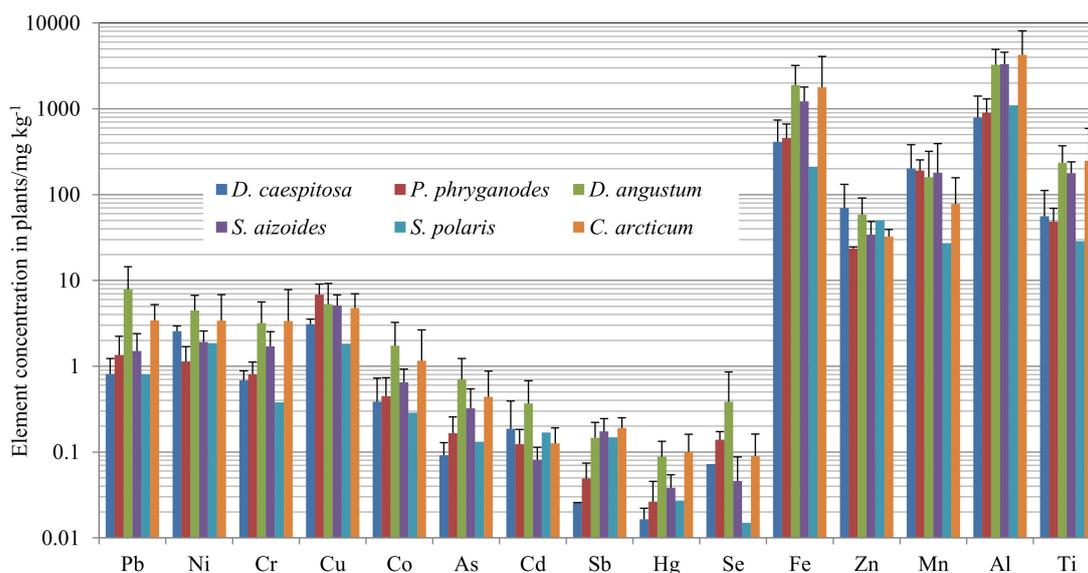
Figure 2. Element concentration means in the six species of plants, with error bars of one standard deviation. The sample numbers of plants *D. caespitosa*, *P. phryganodes*, *D. angustum*, *S. aizoides*, *S. Polaris*, and *C. arcticum* are 2, 3, 6, 4, 1, and 3, respectively. Note that a base-10 log scale is used for the *y*-axis of element concentrations.

4. Discussion

4.1. Spatial Variation of Elements in Plants

According to the distance from settlement points, the sampling stations were divided into two groups, Zones I and II (Figure 1). Zone I includes sites S2, S4, and S5, which are close to the settlement points, research stations, airport, and the main roads. In this case, elements in plants in Zone I tend to be influenced by local human activities, while sampling sites in Zone II (S1, S3, and S6) are away from the residential areas.

Element concentrations in the six species of plants in the two zones are shown in Figure 3, and element means in Zone I are generally higher than those of Zone II, especially Pb, Cd, Se, Co, and Zn. The spatial distribution pattern of elements in *D. angustum* is likely more representative due to its largest sample numbers being among the six plant species (i.e., three samples in Zone I and three samples in Zone II). For *D. angustum*, the means of 10 elements are generally higher in Zone I than in Zone II, with the ratios (Zone I over Zone II) of 2.2(Pb), 1.2(Ni), 1.3(Cu), 1.6(Co), 3.2(Cd), 1.2(Sb), 1.9(Hg), 3.3(Se), 1.1(Zn), and 1.9(Mn). It is noted that element levels in *D. angustum* at S3 are relatively low among the six sites. This is probably due to S3 being far from the settlement point in comparison with other sites (Figure 1) and/or S3 being mainly in the prevailing windward direction of the study region (winds blowing from the sea to the land) [31], and thus less influenced by local human emissions. Interestingly, a previous investigation has reported a similar spatial distribution pattern of elements in soils in Svalbard [46]. It is noted that element contents in plants at S2 are not significantly higher than those of other sites, although S2 is close to a coal mine. It might be suggested that the impacts of coal mining activities before the 1960s on element levels in Svalbard plants are rather minor.
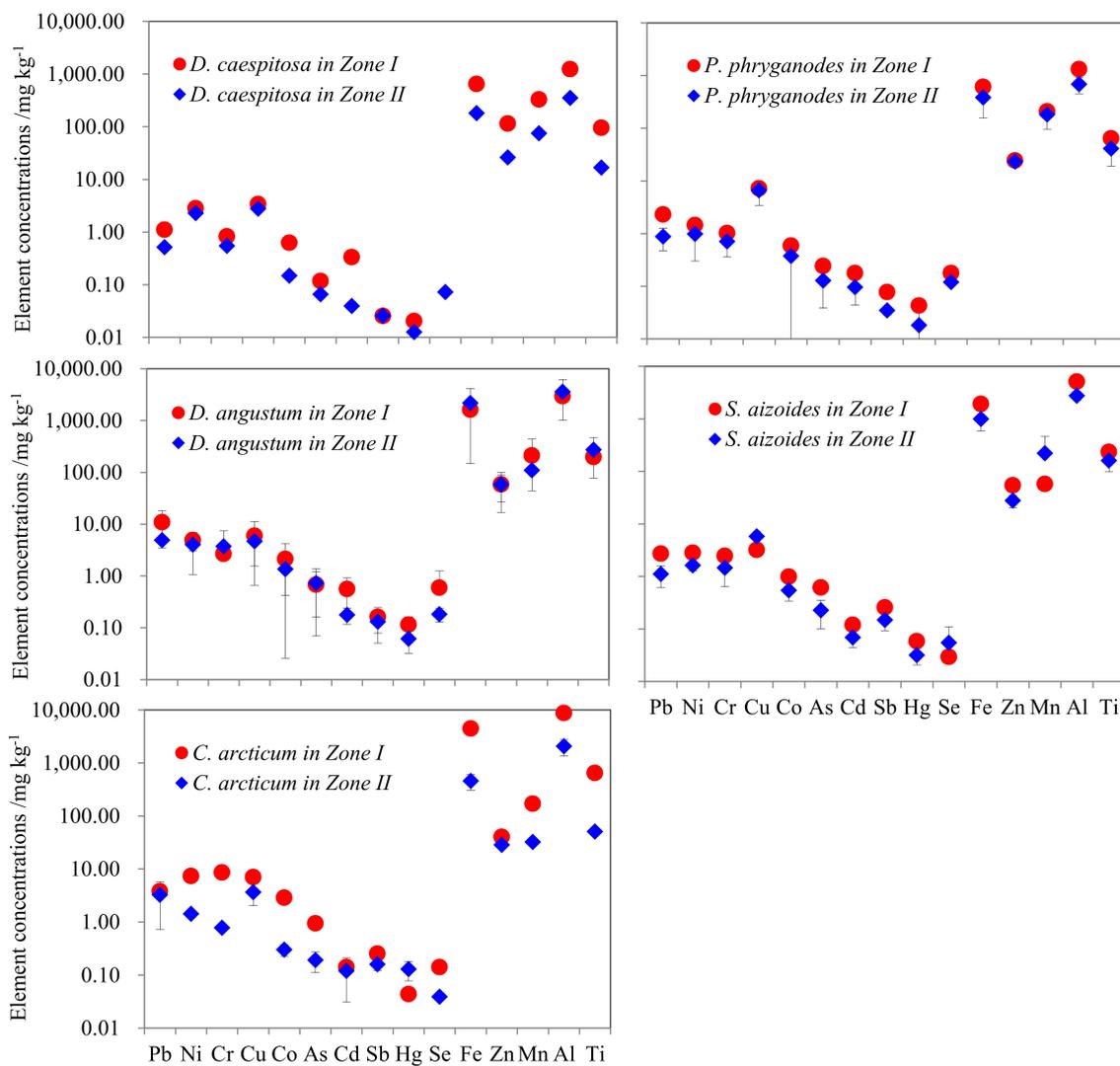
Figure 3. Element concentration means in different plants in the two zones. Note that only one sample of *S. polaris* was collected, and this species was excluded. Error bars represent one standard deviation of element concentrations in individual zones. Note that a base-10 log scale is used for the *y*-axis of element concentrations.

In summary, the spatial distribution patterns of elements possibly suggests that local anthropogenic activities are influential on some elements in plants in Ny-Ålesund. However, it is hard to assess to what extent the local human activities have influenced element levels in plants based on our data.

4.2. Enrichment Factors of Elements in Plants

EFs of elements in six species of plants are illustrated in Figure 4. The six species of plants show a similar EF trend among different elements, generally following a decreasing order of Cd > Hg > Zn > Sb > Mn > Cu > Pb > Ni > As > Co > Ti > Fe > Cr. EFs of Cd, Hg, and Zn in plants are much higher than 10.0, suggesting that these elements were influenced by other sources which may be associated with anthropogenic inputs. Most of the EFs of other elements are below or close to 10.0, suggesting an influence from soil particles. It is noted that EFs of Cr, Co, Fe, and Ti are close to 1.0, possibly indicating that the crustal source plays an important role in accumulation of these metals.
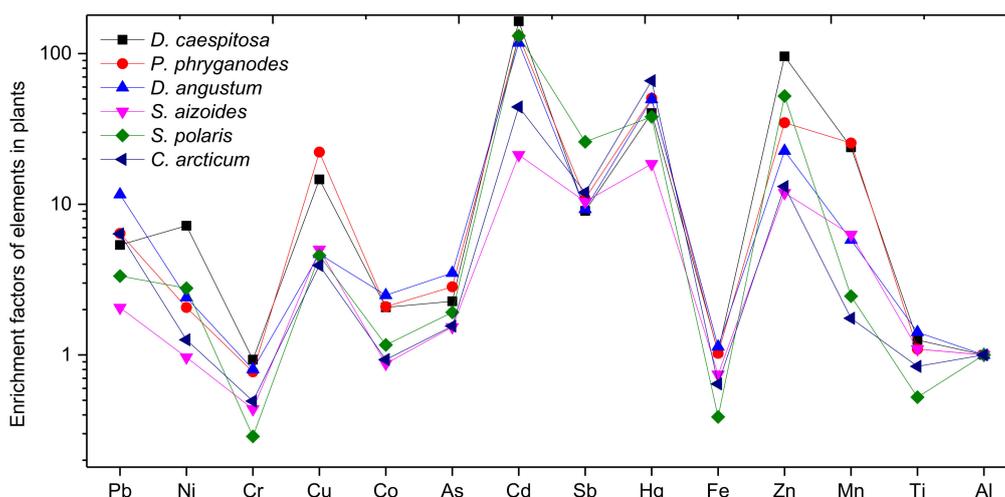
Figure 4. Means of the enrichment factors of elements in the six species of plants in Ny-Ålesund, the Arctic. Note that a base-10 log scale is used for the *y*-axis of enrichment factors of elements.

In terms of different plants, EFs vary moderately among the six species (Figure 4). Different from element concentration patterns of interspecies (Section 3), the EFs in *D. caespitosa* are generally high, while the EFs in *C. arcticum* are relatively low. This difference possibly suggests that both concentrations and EFs of elements should be considered for assessing the suitability of plants to monitor anthropogenic inputs of pollutants. The differences in EFs of elements amongst species could be associated with the different element adsorption capacity of plants [47], assuming that element contents in soils at each site are similar and elements in plants are mainly from local soils. In this study, although there is uncertainty regarding EF estimation (i.e., taking top soils as the reference environment instead of the local root-zone soils), the EFs of elements in plants allow a useful first-order examination of element sources. It is noted that only one *S. Polaris* sample was measured, and again EFs in *S. Polaris* could remain uncertain, especially when compared with other species.

4.3. Element Groups in Plants

Elements in plants can have various sources, including local and long-range origins, absorption of elements from the bedrock and soil, natural cycling process (e.g., sea salts and biogenic emissions), as well as windblown soil dust [18,48]. In order to identify the main factors influencing element levels, three principal components (PCs), with initial eigenvalues >1.0, were extracted using the PCA. The three components account for 82% of the total variance in element concentrations, suggesting that most of the variation of elements in plants can be explained by the three factors. The plot loadings of different elements in three components are shown in Figure 5.
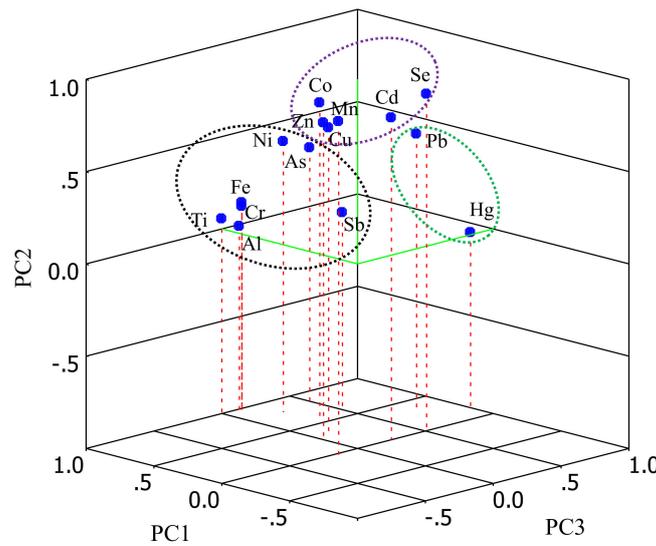
Figure 5. Three-dimensional plot loadings (PC1, PC2, and PC3) of elements in plants by principal component analysis (PCA). Varimax rotation method was applied in factor analysis to highlight the main influence factors. The method of regression was chosen for calculation of factor score coefficient, and three components were extracted.

PC1 could explain 53% of the total variance, in which Ni, Cr, As, Sb, Fe, Al, and Ti are highly loaded, with loadings of 0.79, 0.96, 0.76, 0.58, 0.97, 0.96, and 0.97, respectively. These seven elements are correlated well with each other (Table 3), possibly indicating that a common source controls their concentrations [49–51]. In general, EFs of the seven elements are generally lower than or close to 10.0 (Figure 4), likely suggesting an influence from soil sources. In addition, Fe, Al, and Ti are the typical lithophilic elements, usually selected as the reference elements in enrichment factor analysis [37,38,52]. In this case, PC1 may be interpreted as a mineral particle factor, i.e., the windblown soil dust. This is consistent with previous reports that mineral particle is a main factor influencing Al, Cr, Fe, and Ti contents in plants [18,53,54]. In a previous investigation in Greenland, the first principal component of element concentrations in plants is also interpreted as a soil factor, consistent with our results [35]. Therefore, it seems that windblown mineral particles can account for most of the total variation of elements in plants around the Arctic. It is noted that Co also shows a positive loading in PC1 (0.59), indicating that Co in plants is also influenced by soil dust.

Table 3. Non-parametric Spearman correlation coefficients of element concentrations in plants collected in Ny-Ålesund, the Arctic (*n* = 19).

	Pb	Ni	Cr	Cu	Co	As	Cd	Sb	Hg	Se	Fe	Zn	Mn	Al	Ti
Pb	1.00	0.57 *	0.80 **	0.21	0.67 **	0.84 **	0.48 *	0.51 *	0.85 **	0.62 **	0.70 **	0.26	−0.11	0.65 **	0.61 **
Ni		1.00	0.78 **	0.14	0.81 **	0.73 **	0.36	0.30	0.34	0.47	0.73 **	0.53 *	0.06	0.62 **	0.73 **
Cr			1.00	0.37	0.91 **	0.93 **	0.22	0.48 *	0.58 **	0.58 *	0.92 **	0.22	0.07	0.79 **	0.88 **
Cu				1.00	0.51 *	0.40	−0.07	0.24	0.07	0.42	0.42	0.14	0.58 **	0.24	0.33
Co					1.00	0.86 **	0.33	0.51 *	0.43	0.56 *	0.93 **	0.44	0.25	0.80 **	0.90 **
As						1.00	0.20	0.64 **	0.70 **	0.57 *	0.89 **	0.31	−0.05	0.81 **	0.82 **
Cd							1.00	0.05	0.33	0.49	0.21	0.55 *	0.28	0.16	0.24
Sb								1.00	0.58 **	0.28	0.67 **	0.32	−0.02	0.74 **	0.56 *
Hg									1.0 0	0.61 *	0.53 *	0.22	−0.26	0.60 **	0.43
Se										1.00	0.54 *	0.26	0.58 *	0.31	0.44
Fe											1.00	0.41	0.22	0.91 **	0.96 **
Zn												1.00	0.37	0.40	0.38
Mn													1.00	0.01	0.20
Al														1.00	0.93 **
Ti															1.00

(a) * significant at the 0.05 level (two-tailed). (b) ** significant at the 0.01 level (two-tailed).

PC2 explains 19% of the total variance of element concentrations. Elements Cd, Se, Mn, Co, Cu, and Zn are highly loaded in PC2, with loadings of 0.74, 0.82, 0.82, 0.70, 0.67, and 0.71, respectively. Furthermore, there is a better correlation between these elements (Table 3), possibly suggesting a similar main source. In spatial terms, their concentrations in moss *D. angustum*, the most widely distributed species in the study area (i.e., present at all six sampling sites), are generally higher in Zone I than in Zone II, possibly indicating a local human influence. Moreover, the mean EFs of Cd and Zn are 102 and 38, respectively, much higher than 10.0, suggesting human sources. Thus, PC2 possibly represents an influence from anthropogenic activities. In Ny-Ålesund, the local human activities, such as transportation, operation of the research stations, and residential emissions, could potentially contribute to elements in different environments (Figure 1) [46,55,56]. Moreover, the long-distance atmospheric transport of elements to the Arctic was thought to be an important source [35,57]. The long-range transport, however, does not seem to play a dominant role regarding the significant spatial variation in plant elements. Based upon the available data, it is difficult to distinguish the pollutants associated with long-range atmospheric transport or local point sources.

PC3 explains 10% of the total variance, and is highly correlated with Hg and Pb, possibly indicating a common source of the two metals. In addition, the two metals are correlated well with each other ($r = 0.85$, $p < 0.01$; Table 3), likely suggesting a common source. It is noted that all Hg EFs in plants are much higher than 10.0 (Figure 4); this is most likely associated with the human inputs rather than with soil dust. Among the 15 elements in this study, Hg arouses the greatest concern from researchers, with loadings in Arctic environments appearing to be increasing due to human activities [58,59]. Previous investigations have suggested that long-range transport is responsible for Hg accumulation in the Arctic [35,60]. In *D. angustum*, the highest level of Hg is present at S5, while in *C. arcticum*, the highest value is found at S6 rather than at S5. In this case, it is hard to attribute Hg accumulation in plants only to the local stationary point source. Similarly, Hg-Pb and Cd-Zn in the plants in Greenland were also classified into two principal components (i.e., PC2 and PC3, respectively), and the four elements were thought to be mainly from human sources rather than soil dust [35].

It is noted that the element data in different species were used for the PCA, and the number of samples is relatively small here. Consequently, the outcomes of PCA, at least for a specific species, maintained some uncertainty. Still, the PCA results here can provide the general information on the sources of elements in plants collected in Ny-Ålesund, which will be of significance to a better understanding of element enrichment in the Arctic ecosystem.

5. Conclusions

We have performed an investigation focusing on element accumulation in different plants in Ny-Ålesund, the Arctic, to characterize the enrichment and sources of elements in plants. While few samples were collected from individual species, the measurements provide the general information on element accumulation status in Arctic plants. It is shown that element concentrations vary among different species, with elevated concentrations in *C. arcticum* and *D. angustum*. In spatial, element levels are generally higher in the plants near the settlement points than those of other sites, indicating the local human sources.

The enrichment factor (EF) analysis showed that Cd, Hg, and Zn in plants are significantly enriched with respect to the Spitsbergen topsoil elements, while EFs of Cr, Co, Fe, and Ti are close to 1.0 (i.e., no enrichment). In the six species of plants, EFs of elements in *D. caespitosa* are generally elevated, indicating that *D. caespitosa* could be a more sensitive bioindicator of elements, compared with other species.

With the aid of PCA, the elements in plants were classified into three groups. The elements Ni, Cr, As, Sb, Fe, Al, Ti, and to a lesser extent Co, are mainly influenced by continental crust particles. Therefore, the atmospheric deposition of these elements could not be monitored by the plants. The levels of Cd, Se, Mn, Cu, Zn, Hg, and Pb are likely associated with anthropogenic activities, but it is difficult to distinguish the origins between the local emissions and the long-range atmospheric

transport of pollutants. Together with the high EFs of Cd, Hg, and Zn, it can be concluded that, for these three metals, the plants seem to have a great potential for monitoring the atmospheric transport (i.e., an effective biomonitor) in Ny-Ålesund. Together with the contents of the three metals in plants, the moss *D. angustum* is likely more effective at monitoring the atmospheric deposition of elements in the study area.

References

1. Barrie, L.A. Arctic air pollution: An overview of current knowledge. *Atmos. Environ.* **1986**, *20*, 643–663. [CrossRef]

2. Simões, J.C.; Zagorodnov, V.S. The record of anthropogenic pollution in snow and ice in Svalbard, Norway. *Atmos. Environ.* **2001**, *35*, 403–413. [CrossRef]

3. Zheng, J.; Shotyk, W.; Krachler, M.; Fisher, D.A. A 15,800-year record of atmospheric lead deposition on the Devon Island Ice Cap, Nunavut, Canada: Natural and anthropogenic enrichments, isotopic composition, and predominant sources. *Glob. Biogeochem. Cycles* **2007**, *21*. [CrossRef]

4. Michelutti, N.; Simonetti, A.; Briner, J.P.; Funder, S.; Creaser, R.A.; Wolfe, A.P. Temporal trends of pollution Pb and other metals in east-central Baffin Island inferred from lake sediment geochemistry. *Sci. Total Environ.* **2009**, *407*, 5653–5662. [CrossRef] [PubMed]

5. Liu, X.; Jiang, S.; Zhang, P.; Xu, L. Effect of recent climate change on Arctic Pb pollution: A comparative study of historical records in lake and peat sediments. *Environ. Pollut.* **2012**, *160*, 161–168. [CrossRef] [PubMed]

6. Shi, G.; Teng, J.; Ma, H.; Li, Y.; Sun, B. Metals and metalloids in precipitation collected during CHINARE campaign from Shanghai, China to Zhongshan Station, Antarctica: Spatial variability and source identification. *Glob. Biogeochem. Cycles* **2015**, *29*, 760–774. [CrossRef]

7. Bindler, R.; Renberg, I.; John Anderson, N.; Appleby, P.G.; Emteryd, O.; Boyle, J. Pb isotope ratios of lake sediments in West Greenland: Inferences on pollution sources. *Atmos. Environ.* **2001**, *35*, 4675–4685. [CrossRef]

8. Allen-Gil, S.M.; Ford, J.; Lasorsa, B.K.; Monetti, M.; Vlasova, T.; Landers, D.H. Heavy metal contamination in the Taimyr Peninsula, Siberian Arctic. *Sci. Total Environ.* **2003**, *301*, 119–138. [CrossRef]

9. Fitzgerald, W.F.; Engstrom, D.R.; Lamborg, C.H.; Tseng, C.-M.; Balcom, P.H.; Hammerschmidt, C.R. Modern and historic atmospheric mercury fluxes in northern Alaska: Global sources and Arctic depletion. *Environ. Sci. Technol.* **2005**, *39*, 557–568. [CrossRef]

10. Shotyk, W.; Zheng, J.; Krachler, M.; Zdanowicz, C.; Koerner, R.; Fisher, D. Predominance of industrial Pb in recent snow (1994–2004) and ice (1842–1996) from Devon Island, Arctic Canada. *Geophys. Res. Lett.* **2005**, *32*. [CrossRef]

11. Lahoutifard, N.; Sparling, M.; Lean, D. Total and methyl mercury patterns in Arctic snow during springtime at Resolute, Nunavut, Canada. *Atmos. Environ.* **2005**, *39*, 7597–7606. [CrossRef]

12. Cai, M.; Lin, J.; Hong, Q.; Wang, Y.; Cai, M. Content and distribution of trace metals in surface sediments from the northern Bering Sea, Chukchi Sea and adjacent Arctic areas. *Mar. Pollut. Bull.* **2011**, *63*, 523–527. [CrossRef] [PubMed]

13. Fort, J.; Grémillet, D.; Traisnel, G.; Amélineau, F.; Bustamante, P. Does temporal variation of mercury levels in Arctic seabirds reflect changes in global environmental contamination, or a modification of Arctic marine food web functioning? *Environ. Pollut.* **2016**, *211*, 382–388. [CrossRef] [PubMed]

14. Dietz, R.; Sonne, C.; Basu, N.; Braune, B.; O'Hara, T.; Letcher, R.J.; Scheuhammer, T.; Andersen, M.; Andreasen, C.; Andriashek, D. What are the toxicological effects of mercury in Arctic biota? *Sci. Total Environ.* **2013**, *443*, 775–790. [CrossRef] [PubMed]

15. Hansson, S.V.; Desforges, J.-P.; van Beest, F.M.; Bach, L.; Halden, N.M.; Sonne, C.; Mosbech, A.; Søndergaard, J. Bioaccumulation of mining derived metals in blood, liver, muscle and otoliths of two Arctic predatory fish species (Gadus ogac and Myoxocephalus scorpius). *Environ. Res.* **2020**, *183*, 109194. [CrossRef]

16. Kicińska, A.; Gruszecka-Kosowska, A. Long-term changes of metal contents in two metallophyte species (Olkusz area of Zn-Pb ores, Poland). *Environ. Monit. Assess* **2016**, *188*, 339. [CrossRef]

17. Kicińska, A. Assessment of the road traffic impact on accumulation of selected elements in soils developed on Krynica and Bystrica subunit (Magura Nappe, Polish Outer Carpathians). *Carpathian J. Earth Environ. Sci.* **2016**, *11*, 245–254.

18. Szczepaniak, K.; Biziuk, M. Aspects of the biomonitoring studies using mosses and lichens as indicators of metal pollution. *Environ. Res.* **2003**, *93*, 221–230. [CrossRef]

19. Grodzińska, K.; Szarek-Łukaszewska, G. Response of mosses to the heavy metal deposition in Poland-an overview. *Environ. Pollut.* **2001**, *114*, 443–451. [CrossRef]

20. Zechmeister, H.; Hohenwallner, D.; Riss, A.; Hanus-Illnar, A. Variations in heavy metal concentrations in the moss species *Abietinella abietina* (Hedw.) Fleisch. according to sampling time, within site variability and increase in biomass. *Sci. Total Environ.* **2003**, *301*, 55–65. [CrossRef]

21. Harmens, H.; Buse, A.; Büker, P.; Norris, D.; Mills, G.; Williams, B.; Reynolds, B.; Ashenden, T.W.; Rühling, Å.; Steinnes, E. Heavy metal concentrations in European mosses: 2000/2001 survey. *J. Atmos. Chem.* **2004**, *49*, 425–436. [CrossRef]

22. Kłos, A.; Ziembik, Z.; Rajfur, M.; Dołhańczuk-Śródka, A.; Bochenek, Z.; Bjerke, J.W.; Tømmervik, H.; Zagajewski, B.; Ziółkowski, D.; Jerz, D.; et al. Using moss and lichens in biomonitoring of heavy-metal contamination of forest areas in southern and north-eastern Poland. *Sci. Total Environ.* **2018**, *627*, 438–449. [CrossRef]

23. Reimann, C.; Niskavaara, H.; Kashulina, G.; Filzmoser, P.; Boyd, R.; Volden, T.; Tomilina, O.; Bogatyrev, I. Critical remarks on the use of terrestrial moss (*Hylocomium splendens* and *Pleurozium schreberi*) for monitoring of airborne pollution. *Environ. Pollut.* **2001**, *113*, 41–57. [CrossRef]

24. Buse, A.; Norris, D.; Harmens, H.; Büker, P.; Ashenden, T.; Mills, G. *Heavy Metals in European Mosses: 2000/2001 Survey*; ICP Vegetation Programme Coordination Centre, Centre for Ecology and Hydrology: Bangor, UK, 2003; Available online: http://icpvegetation.ceh.ac.uk (accessed on 19 October 2020).

25. Harmens, H.; Norris, D.; Steinnes, E.; Kubin, E.; Piispanen, J.; Alber, R.; Aleksiayenak, Y.; Blum, O.; Coşkung, M.; Dam, M. Mosses as biomonitors of atmospheric heavy metal deposition: Spatial patterns and temporal trends in Europe. *Environ. Pollut.* **2010**, *158*, 3144–3156. [CrossRef] [PubMed]

26. Schnyder, E.; Štrok, M.; Kosonen, Z.; Skudnik, M.; Mazej, D.; Jeran, Z.; Thöni, L. Lead concentrations and stable lead isotope ratios in moss in Slovenia and Switzerland. *Ecol. Indic.* **2018**, *95*, 250–259. [CrossRef]

27. Wojtuń, B.; Samecka-Cymerman, A.; Kolon, K.; Kempers, A.J.; Skrzypek, G. Metals in some dominant vascular plants, mosses, lichens, algae, and the biological soil crust in various types of terrestrial tundra, SW Spitsbergen, Norway. *Polar. Biol.* **2013**, *36*, 1799–1809. [CrossRef]

28. Jia, N.; Sun, L.; He, X.; You, K.; Zhou, X.; Long, N. Distributions and impact factors of antimony in topsoils and moss in Ny-Ålesund, Arctic. *Environ. Pollut.* **2012**, *171*, 72–77. [CrossRef]

29. Hisdal, V.; Polarinstitutt, N. *Svalbard: Nature and History*; Norsk Polarinstitutt: Oslo, Norway, 1998.

30. Jiang, S.; Liu, X.; Sun, J.; Yuan, L.; Sun, L.; Wang, Y. A multi-proxy sediment record of late Holocene and recent climate change from a lake near Ny-Ålesund Svalbard. *Boreas* **2011**, *40*, 468–480. [CrossRef]

31. Beine, H.J.; Argentini, S.; Maurizi, A.; Mastrantonio, G.; Viola, A. The local wind field at Ny-Ålesund and the Zeppelin mountain at Svalbard. *Meteorol. Atmos. Phys.* **2001**, *78*, 107–113. [CrossRef]

32. Aboal, J.; Pérez-Llamazares, A.; Carballeira, A.; Giordano, S.; Fernández, J. Should moss samples used as biomonitors of atmospheric contamination be washed? *Atmos. Environ.* **2011**, *45*, 6837–6840. [CrossRef]

33. Markert, B.; Wappelhorst, O.; Weckert, V.; Herpin, U.; Siewers, U.; Friese, K.; Breulmann, G. The use of bioindicators for monitoring the heavy-metal status of the environment. *J. Radioanal. Nucl. Chem.* **1999**, *240*, 425–429. [CrossRef]

34. Migaszewski, Z.M.; Gałuszka, A.; Pasławski, P. Polynuclear aromatic hydrocarbons, phenols, and trace metals in selected soil profiles and plant bioindicators in the Holy Cross Mountains, South-Central Poland. *Environ. Int.* **2002**, *28*, 303–313. [CrossRef]

35. Riget, F.; Asmund, G.; Aastrup, P. The use of lichen (*Cetraria nivalis*) and moss (*Rhacomitrium lanuginosum*) as monitors for atmospheric deposition in Greenland. *Sci. Total Environ.* **2000**, *245*, 137–148. [CrossRef]

36. Aceto, M.; Abollino, O.; Conca, R.; Malandrino, M.; Mentasti, E.; Sarzanini, C. The use of mosses as environmental metal pollution indicators. *Chemosphere* **2003**, *50*, 333–342. [CrossRef]

37. Turner, A.; Simmonds, L. Elemental concentrations and metal bioaccessibility in UK household dust. *Sci. Total Environ.* **2006**, *371*, 74–81. [CrossRef]

38. Meza-Figueroa, D. Heavy metal distribution in dust from elementary schools in Hermosillo, Sonora, México. *Atmos. Environ.* **2007**, *41*, 276–288. [CrossRef]

39. Shi, G.; Li, Y.; Jiang, S.; An, C.; Ma, H.; Sun, B.; Wang, Y. Large-scale spatial variability of major ions in the atmospheric wet deposition along the China Antarctica transect (31°N~69°S). *Tellus B* **2012**, *64*, 17134. [CrossRef]

40. Lawrence, C.R.; Neff, J.C. The contemporary physical and chemical flux of aeolian dust: A synthesis of direct measurements of dust deposition. *Chem. Geol.* **2009**, *267*, 46–63. [CrossRef]

41. Jiang, S.Y.N.; Yang, F.; Chan, K.L.; Ning, Z. Water solubility of metals in coarse PM and $PM_{2.5}$ in typical urban environment in Hong Kong. *Atmos. Pollut. Res.* **2014**, *5*, 236–244. [CrossRef]

42. Rudnick, R.; Gao, S. Composition of the continental crust. *Treatise Geochem.* **2003**, *3*, 1–64.

43. Chiarenzelli, J.R.; Aspler, L.B.; Ozarko, D.L.; Hall, G.; Powis, K.B.; Donaldson, J. Heavy metals in lichens, southern district of Keewatin, Northwest Territories, Canada. *Chemosphere* **1997**, *35*, 1329–1341. [CrossRef]

44. France, R.; Coquery, M. Lead concentrations in lichens from the Canadian high arctic in relation to the latitudinal pollution gradient. *Water Air Soil Pollut.* **1996**, *90*, 469–474. [CrossRef]

45. Grodzińska, K.; Godzik, B. Heavy metals and sulphur in mosses from southern Spitsbergen. *Polar. Res.* **1991**, *9*, 133–140. [CrossRef]

46. Gulińska, J.; Rachlewicz, G.; Szczuciński, W.; Barałkiewicz, D.; Kózka, M.; Bulska, E.; Burzyk, M. Soil contamination in high Arctic areas of human impact, central Spitsbergen, Svalbard. *Pol. J. Environ. Stud.* **2003**, *12*, 701–707.

47. Koz, B.; Cevik, U. Lead adsorption capacity of some moss species used for heavy metal analysis. *Ecol. Indic.* **2014**, *36*, 491–494. [CrossRef]

48. Lajayer, B.A.; Moghadam, N.K.; Maghsoodi, M.R.; Ghorbanpour, M.; Kariman, K. Phytoextraction of heavy metals from contaminated soil, water and atmosphere using ornamental plants: Mechanisms and efficiency improvement strategies. *Environ. Sci. Pollut. Res.* **2019**, *26*, 8468–8484. [CrossRef] [PubMed]

49. Shi, G.; Chen, Z.; Xu, S.; Zhang, J.; Wang, L.; Bi, C.; Teng, J. Potentially toxic metal contamination of urban soils and roadside dust in Shanghai, China. *Environ. Pollut.* **2008**, *156*, 251–260. [CrossRef]

50. Wolterbeek, B. Biomonitoring of trace element air pollution: Principles, possibilities and perspectives. *Environ. Pollut.* **2002**, *120*, 11–21. [CrossRef]

51. Wolterbeek, H.T.; Verburg, T. Judging survey quality: Local variances. *Environ. Monit. Assess* **2002**, *73*, 7–16. [CrossRef]

52. Shi, G.; Chen, Z.; Teng, J.; Bi, C.; Zhou, D.; Sun, C.; Li, Y.; Xu, S. Fluxes, variability and sources of cadmium, lead, arsenic and mercury in dry atmospheric depositions in urban, suburban and rural areas. *Environ. Res.* **2012**, *113*, 28–32. [CrossRef]

53. Berg, T.; Steinnes, E. Recent trends in atmospheric deposition of trace elements in Norway as evident from the 1995 moss survey. *Sci. Total Environ.* **1997**, *208*, 197–206. [CrossRef]

54. Steinnes, E. A critical evaluation of the use of naturally growing moss to monitor the deposition of atmospheric metals. *Sci. Total Environ.* **1995**, *160*, 243–249. [CrossRef]

55. Hao, Z.L.; Wang, F.; Yang, H.Z. Baseline values for heavy metals in soils on Ny-Ålesund, Spitsbergen Island, Arctic: The extent of anthropogenic pollution. *Adv. Mater. Res.* **2013**, *779–780*, 1260–1265. [CrossRef]

56. Poland, J.S.; Riddle, M.J.; Zeeb, B.A. Contaminants in the Arctic and the Antarctic: A comparison of sources, impacts, and remediation options. *Polar Rec.* **2003**, *39*, 369. [CrossRef]

57. Singh, D.K.; Kawamura, K.; Yanase, A.; Barrie, L.A. Distributions of polycyclic aromatic hydrocarbons, aromatic ketones, carboxylic acids, and trace metals in arctic aerosols: Long-range atmospheric transport, photochemical degradation/production at polar sunrise. *Environ. Sci. Technol.* **2017**, *51*, 8992–9004. [CrossRef]

58. Macdonald, R.; Barrie, L.; Bidleman, T.; Diamond, M.; Gregor, D.; Semkin, R.; Strachan, W.; Li, Y.; Wania, F.; Alaee, M. Contaminants in the Canadian Arctic: 5 years of progress in understanding sources, occurrence and pathways. *Sci. Total Environ.* **2000**, *254*, 93–234. [CrossRef]

59. Halbach, K.; Mikkelsen, Ø.; Berg, T.; Steinnes, E. The presence of mercury and other trace metals in surface soils in the Norwegian Arctic. *Chemosphere* **2017**, *188*, 567–574. [CrossRef]

60. Obrist, D.; Agnan, Y.; Jiskra, M.; Olson, C.L.; Colegrove, D.P.; Hueber, J.; Moore, C.W.; Sonke, J.E.; Helmig, D. Tundra uptake of atmospheric elemental mercury drives Arctic mercury pollution. *Nature* **2017**, *547*, 201–204. [CrossRef]