Research Article

Application of Neutron Activation Analysis for Determination of As, Cr, Hg, and Se in Mosses in the Metropolitan Area of the Valley of Toluca, Mexico

R. Mejía-Cuero,^{1,2} G. García-Rosales,² L. C. Longoria-Gándara,^{1,3}
M. C. López-Reyes,¹ and P. Ávila-Pérez^{1,2}

¹National Institute for Nuclear Research, Carretera México-Toluca S/N, La Marquesa, 52750 Ocoyoacac, MEX, Mexico

²Departamento de Posgrado, Instituto Tecnológico de Toluca, Ex-Rancho la Virgen S/N, 50120 Metepec, MEX, Mexico

³Division for Latin America, Department of Technical Cooperation, International Atomic Energy Agency, Wagramer Strasse 5, P.O. Box 100, 1400 Vienna, Austria

Correspondence should be addressed to G. García-Rosales; ggarciarosales508@gmail.com

Received 12 December 2014; Revised 26 March 2015; Accepted 23 April 2015

Academic Editor: Qiang Jin

Copyright © 2015 R. Mejía-Cuero et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

This research presents a study of environmental monitoring at different sampling sites from the Metropolitan Area of the Valley of Toluca (MAVT), Mexico, using mosses (*Leskea angustata* (Tayl.) and *Fabronia ciliaris* (Brid.)) and soil samples. The epiphytic mosses and soils were sampled in two campaigns within two periods of the year, a rainy and dry-cold season. The selected sampling sites included urban regions (UR), transitional regions (TR), and protected natural areas (PA). The samples were analyzed by the Instrumental Neutron Activation Analysis (INAA) to determine As, Cr, Hg, and Se principally. However, due to the versatility of the analytic technique used, other elements including Cs, Co, Sc, Sb, Rb, Ce, La, Eu, and Yb were also detected. Statistical analysis (As, Cr, Hg, and Se) was carried out with principal components and cluster analysis methods; this revealed that a good correlation exists between metal content in mosses and the degree of pollution in the areas sampled. The obtained results in mosses showed that the concentrations of As, Cr, Co, Cs, Rb, Ce, La, and Yb increased with respect to the concentrations obtained during the first sampling, whereas Se, Sc, Sb and Eu, concentrations were decreased. For As and Hg, the concentrations were similar in both sampling periods. The soil samples present the most significant concentration.

1. Introduction

The Metropolitan Area of the Valley of Toluca (MAVT) is located in the central area of the State of Mexico and is comprised of seven municipalities with a population of about 1.8 million. It is the most industrialized region of Mexico with different areas of industry (tanneries, electroplating, textile, and kraft pulp bleaching) and also the significant amount of traffic [1], resulting in the emission of various pollutants into the atmosphere. Currently the deposition levels of metals including As, Cr, Hg, and Se from the atmosphere to the biosphere may be significantly increased as a result of the anthropogenic input of fossil fuels, dust from agriculture, industry and metallurgy, or natural sources. Due to the diversity of their habitats, their structural simplicity, and

rapid rate of multiplication, some mosses may be useful tools for prospective studies to determine environmental conditions and are ideal organisms for studying deposition of pollutants from the atmosphere to vegetation [2]. Mosses are found in many different environments and considered as indicators of elemental pollution [3]. Several previous studies have used plants and mosses as biomonitors because they facilitate the measurement of pollutant deposition [4]. The diversity of mosses depends on the weather and substrate where they develop [5] and the factors limiting their distribution are essentially water, temperature, and altitude. The concentration of metals and other elements in mosses may depend on morphological features [6], wind direction, topography, altitude, latitude, and time of exposure. Additionally, topsoil analysis has been widely used to evaluate

the uptake of contaminants in ecosystems, to estimate the maximum concentration of metal and other elements, and to evaluate their importance as a source of the metals and other elements absorbed by mosses [7]. The organic fraction of the topsoil, in particular, the humus, can be used for measuring atmospheric deposition of metals and other elements [8]. In this context, the aim of this research is to determine the concentrations of As, Cr, Hg, and Se in mosses and topsoil from 16 sampling sites in MAVT using INAA. The advantage of using INAA is the minimal sample preparation needed for analysis compared to methods and its multielement that can be nondestructive, with adequate limits of detection for the majority of metals and other elements of environmental interest [9].

2. Material and Methods

2.1. Sampling. First of all, the sampling sites were carefully inspected to determine the moss species present at each site (7 different moss species were identified). However, not all were found in all the sites investigated due to the characteristics of each area. Consequently, the main two dominant species of epiphytic mosses were *Fabronia ciliaris* (Brid.) and *Leskea angustata* (Tayl.); these were collected in two sampling campaigns from the urban region (UR), transitional region (TR), and the protected natural area (PA). The first sampling was performed in November 2010 (dry season, autumn) and the second in August 2011 (rainy season, summer); each of the sampling sites was selected after considering the prevailing winds, the proximity of the site compared to areas with higher population density, vehicular traffic, industrial activity, and the availability of mosses.

Ten grams of epiphytic mosses was collected from six to ten trees in a height greater than one meter. Moss samples were removed from the tree using a plastic spatula and placed in polythene bags for transport to the laboratory. During sampling, observations of habitat type and relative density of moss carpets were performed.

For the first sampling, seven sampling points were located in parks in urban regions (UR) (sites 1–7 were identified as Negrete, Alameda, Reforma, Hípico, Pilita, Santín, and Tolloacán), two sampling points (sites 8 and 11, identified as Lomas and San Miguel) were located in transition regions (TR), and three sampling sites were located within natural areas (PA) (sites 13, 15, and 16, identified as San Antonio, Cacalomacán, and Ciervita) (Figure 1). For the second sampling, four sampling points were added to expand the monitored area: three were situated in transition regions (TR) (sites 9, 10, and 12, identified as Acapulco, Pedregal, and Ameyalco) and one was situated in a natural area (PA) (site 14, identified as San Diego). The topsoil samples were collected from the same locations according to previously published methodology [10].

2.2. Sample Preparation. In the laboratory the mosses were placed in trays and dried at room temperature via exposure to sunlight for 5 to 8 days. Then, the samples were ground with an agate mortar and pestle to obtain a particle size of

0.85 mm, and the product was stored in labeled polyethylene bottles (high density). The collected topsoil samples were cleaned from extraneous plant materials and dried at ambient room temperature, sorted to remove gravel, and then homogenized and passed through a stainless-steel sieve of 0.075 mm aperture. The resultant samples were stored in polyethylene bottles (high density).

2.3. Chemical Composition. The INAA was carried out at the Department of Reactor, Neutron Activation Analysis Laboratory, National Institute of Nuclear Research in Mexico (ININ). INAA was conducted using the standard method of analytical procedures and employed, described in detail by Travesi, 1975 [11].

To provide quality control, contents of elements yielding short- and long-lived isotopes were determined using certified standard reference materials (SRM). For analysis of the moss samples, standards were used: Lichen-336 from IAEA (International Atomic Energy Agency), Citrus Leaves SRM-1572 from the US NIST (National Institute of Standards and Technology), and the SRM for topsoil Soil-7 from IAEA and SRM-2711 Montana II Soil from US NIST for the measured elements; those reference materials were analyzed in triplicate along with the survey of moss and topsoil samples. The results for the SRM were within 90–105% of certified values; 40 mg of moss, topsoil, and control was added to quartz ampoules of 9 mm in diameter and 6 cm of length. SRM and samples were irradiated at the TRIGA-MARK III nuclear research reactor at the ININ in Mexico, using a neutron flux density of $0.9 \times 10^{13} \text{ n cm}^{-2} \text{ s}^{-1}$ for 20 hours in a SIFCA position.

After irradiation, the samples were repacked and measured after 6 days for 30 minutes to determine ^{76}As , ^{140}La , ^{175}Yb , and ^{177}Lu , secondly they were measured after 30 days for 3 hours to determine ^{203}Hg , ^{141}Ce , ^{60}Co , ^{51}Cr , ^{86}Rb , ^{124}Sb , and ^{46}Sc , and finally they were measured between 80 and 160 days for 10–16 hours to determine ^{75}Se , ^{152}Eu , and ^{134}Cs . The gamma spectra of the samples were measured with a gamma spectrometer with a HPGe detector at a resolution (FWHM) of 1.9 keV and for the peak at 1332 keV corresponding to ^{60}Co . A multichannel analyzer, the 8192 ORTEC, operated with a peak determination program which was used for analysis. Data processing was performed using the software developed in ININ and Hypermet-PC. The element contents were determined on the basis of SRM and flux comparators [12, 13].

2.4. Statistical Analysis. The correlations between the sampling sites and metal concentration (for Hg, Se, Cr, and As) in mosses and topsoils were determined via statistical analysis with the Statistics 7.0 informatic program [14], using principal component analysis (PCA) with the cluster option (CA).

3. Results and Discussion

3.1. Chemical Composition Analysis in Moss Samples. The results obtained for the first sampling are summarized in Table 1, where it is observed that the presence of As, Cr, Hg, and Se was due to the sensitivity of the analysis technique

FIGURE 1: Sampling sites, wind directions, and altitudes in the MAVT.

used. Other elements such as Co, Cs, Sc, Sb, Rb, Ce, La, Eu, and Yb were also detected. It is important to consider that some of these elements can be present in the plant's tissues as a component of irregularly shaped particles adsorbed to the plant surface.

The results obtained indicate the relative concentrations as $\text{Co} > \text{Cr} > \text{Rb} > \text{Ce} > \text{La} > \text{Sc} > \text{As} > \text{Sb} > \text{Se} > \text{Cs} > \text{Yb} > \text{Hg} > \text{Eu}$. The sites with the highest elemental concentrations are located in UR and TR and they do not demonstrate a trend. This behavior can perhaps be attributed to the geographical site location (topography) and characteristics of each sampling site. The presence of some elements in the samples may be due to natural, local, or secondary sources.

The highest concentrations of As were found in Reforma and Toluacán located in the center of MAVT corresponding to UR and San Antonio located in PA. Arsenic is widely

distributed in soils, water, and air. It is a component of more than 100 different minerals. Their speciation of the element is a key factor in controlling mobility, availability, and toxicity in natural environments. Arsenic occurrence and mobilization take place through a combination of natural processes, for example, through water reactions, biological activity, and volcanic emissions. Anthropogenic activities account for widespread As contamination arising from a variety of industrial processes such as wood preservatives, paints, alloys, semiconductors, fossil fuel combustion, mine wastes, smelting, landfilling, sewerage, and agricultural applications (pesticides and fertilizer) which may also introduce As into the environment [15].

The sites with the high concentrations of Cr are common in Alameda, Toluacán, Hípico, and Negrete, which are located

TABLE 1: Element concentration (mg kg^{-1}) in moss from the first sampling.

Sampling sites	As	Cr	Hg	Se	Co	Cs	Sc	Sb	Rb	Ce	Eu	La	Yb
Negrete (UR)	2.12 ± 0.16	40.70 ± 2.68	0.21 ± 0.06	1.52 ± 0.28	2.66 ± 0.15	$1.43 \pm 0.17^*$	4.08 ± 0.16	1.10 ± 0.22	26.50 ± 1.41	5.61 ± 0.58	$0.400 \pm 0.11^*$	3.43 ± 0.12	0.17 ± 0.04
Alameda (UR)	2.43 ± 0.16	$51.09 \pm 3.37^*$	0.27 ± 0.07	$1.80 \pm 0.34^*$	2.55 ± 0.14	1.23 ± 0.15	4.29 ± 0.16	$2.92 \pm 0.59^*$	23.75 ± 1.34	7.04 ± 1.20	$0.402 \pm 0.11^*$	4.30 ± 0.15	0.50 ± 0.11
Reforma (UR)	3.69 ± 0.30	39.54 ± 3.14	0.36 ± 0.09	0.86 ± 0.17	2.01 ± 0.10	0.67 ± 0.11	2.32 ± 0.09	0.67 ± 0.11	14.61 ± 1.45	4.69 ± 0.45	0.24 ± 0.07	2.71 ± 0.09	0.14 ± 0.03
Hípico (UR)	1.77 ± 0.13	43.33 ± 2.82	0.28 ± 0.07	1.34 ± 0.25	1.74 ± 0.09	0.72 ± 0.09	2.60 ± 0.10	2.27 ± 0.46	19.74 ± 1.15	4.33 ± 0.54	0.15 ± 0.05	2.40 ± 0.09	0.12 ± 0.03
Pilita (UR)	2.25 ± 0.12	28.26 ± 1.78	$0.44 \pm 0.08^*$	1.38 ± 0.15	2.30 ± 0.12	0.86 ± 0.11	2.96 ± 0.12	1.00 ± 0.13	27.38 ± 2.03	10.31 ± 0.85	0.22 ± 0.06	6.20 ± 0.22	0.46 ± 0.11
Santín (UR)	1.52 ± 0.12	28.35 ± 1.77	0.27 ± 0.09	1.33 ± 0.16	4.50 ± 0.22	1.18 ± 0.13	$4.90 \pm 0.20^*$	0.77 ± 0.10	$41.60 \pm 2.90^*$	$13.85 \pm 1.13^*$	0.29 ± 0.08	$8.07 \pm 0.26^*$	$0.69 \pm 0.17^*$
Tolloacán (UR)	$4.87 \pm 0.40^*$	43.97 ± 3.50	0.32 ± 0.08	1.06 ± 0.21	2.78 ± 0.14	0.91 ± 0.14	3.45 ± 0.14	0.84 ± 0.14	18.81 ± 1.84	7.01 ± 0.67	0.39 ± 0.11	3.65 ± 0.14	0.20 ± 0.05
Lomas (TR)	1.07 ± 0.07	13.38 ± 0.82	0.13 ± 0.03	0.85 ± 0.13	1.09 ± 0.06	0.29 ± 0.05	1.16 ± 0.04	0.53 ± 0.10	8.49 ± 0.75	2.03 ± 0.24	0.11 ± 0.029	1.32 ± 0.05	0.08 ± 0.02
San Miguel (TR)	1.73 ± 0.11	29.49 ± 1.82	0.28 ± 0.01	1.26 ± 0.26	2.81 ± 0.14	0.75 ± 0.10	2.95 ± 0.12	1.21 ± 0.16	27.08 ± 1.88	10.71 ± 0.87	0.22 ± 0.06	6.60 ± 0.21	0.50 ± 0.12
San Antonio (PA)	3.25 ± 0.22	16.34 ± 1.31	0.36 ± 0.08	0.69 ± 0.14	1.75 ± 0.09	0.91 ± 0.13	1.78 ± 0.07	0.24 ± 0.04	25.51 ± 2.28	3.20 ± 0.34	0.19 ± 0.05	1.97 ± 0.07	0.09 ± 0.02
Cacalomaacán (PA)	1.13 ± 0.10	17.02 ± 1.12	0.21 ± 0.06	0.91 ± 0.17	1.52 ± 0.08	0.61 ± 0.08	1.99 ± 0.08	0.35 ± 0.08	13.03 ± 0.76	3.49 ± 0.35	0.20 ± 0.06	1.97 ± 0.07	0.12 ± 0.03
Ciervita (PA)	0.95 ± 0.09	11.31 ± 0.73	0.25 ± 0.05	0.93 ± 0.16	$56.82 \pm 0.87^*$	0.83 ± 0.15	1.24 ± 0.05	0.44 ± 0.11	21.04 ± 2.64	2.33 ± 0.26	0.11 ± 0.029	1.26 ± 0.05	0.07 ± 0.02

*Higher concentration.

in UR. These concentrations could be associated with emissions from local point sources [16].

Samples from UR (Pilita, Reforma, and Tollocán) and PA (San Antonio) presented the most significant Hg contamination levels; this element is considered a typical anthropogenic pollutant. The most important anthropogenic sources of Hg are industrial, domestic coal combustion and waste incineration. Hg is known to form in a complex way with organic matter such as methylate and demonstrates higher volatility compared to other metals. These different chemical properties are at least partially responsible for the presence of Hg in airborne atmospheric particulate matter [17]. On the other hand, Hg-bearing rock formations may also contribute to enrichment of Hg in mosses [18].

The samples with the highest concentrations of Se are of UR (Alameda, Negrete, and Pilita). All of these sampling points are located in the central region of the MAVT and are exposed to air currents and the influence of winds, which may enable movement of Se from the earth's crust. This element is a typical tracer for urban air pollution (motor vehicles, oil burning, coal combustion, waste incineration, and others) [2].

The highest Co concentrations were observed in samples from Ciervita (PA), a volcanic zone, where igneous rocks contain a high concentration of this element abound. Analysis of the samples from the first collection demonstrated that the highest concentrations of Cs, Sb, and Sc were found in Negrete, Alameda, and Santín (UR), respectively.

Recently, Sb was reported to be the single most highly enriched element in urban dust. This element poses a significant risk to the environment and human health [19]. Its use is provided in the making of automobile brake pads, plastics, and flame retardants. At present, the brake pads in cars are thought to be the main source of atmospheric Sb, whereas other sources such as burning coal, metallurgy, and waste incineration contribute to the increase of the concentration.

The sites with the highest concentrations of Rb (Santín (UR), Pilita (UR), Negrete (UR), and San Miguel (TR)) and La were Santín (UR), Negrete (UR), and San Miguel (TR). For Ce, Eu, and Yb, Alameda (UR), Santín (UR), San Miguel (TR), Negrete (UR), Tollocán (UR), and Pilita (UR) present the highest concentrations. The presence of rare earth elements could be attributed to the local sedimentary rocks, when the relative concentrations are $Ce > La > Sm$ [20]. In this study, only Ce and La were measured, but this trend is observed in the biomonitoring data, indicating that lanthanide contents in samples were entirely natural.

Also it was observed that, in general, the sites located in TR contained the lowest concentrations compared with values obtained in UR, an exception of Co that presented high concentrations in Ciervita (PA). With some variation in the concentrations measured, this observation is consistent with similar results reported by Zarazúa-Ortega et al., 2013 [21], and Caballero-Segura et al., 2014 [22], who found the highest chemical concentrations in sampled mosses located in UR regions. The elemental chemical concentration variation in the samples could be largely attributed to the combustion processes from different socioeconomic activities and the number of vehicles in use locally. Other factors such as

altitude, wind direction, and the number of anthropogenic activities in each region can contribute to the higher observed concentrations in samples from UR and TR.

The factors affecting the accumulation of pollutants in mosses in the MAVT can be attributed to the change in wind direction and the altitude, because the first sampling was conducted in autumn (November); this region was subject to trade winds; these trade winds demonstrated a weak intensity and predominantly blew from south to north, with a slight clockwise curvature that prevented circulation, resulting in a change in their trajectory (Figure 1) [23]. The altitude and mountainous terrain of the MAVT can influence the accumulation of pollutants in specific regions. Recent studies by Gerdol et al., 2002 [24], revealed that altitude is an important factor that influences the deposition of contaminants in mosses.

In the results of the second sampling, presented in Table 2, it is observed that the relative order of pollutant accumulation was $Cr > Rb > Ce > La > Co > Sc > Sb > Cs > Se > As > Eu > Hg >$ and Yb . It is shown that the highest concentrations are located in UR and TR regions.

A comparison of the results obtained from the first and second sampling events (Figure 2) demonstrated that in general the analyzed samples from the first sampling event (Table 1) contained elemental concentrations greater than those measured in samples from the second sampling. The increase in concentration can be attributed to the effect of the rainy season and anthropogenic activities in the MAVT. If the summer season is rainy, it could increase elemental concentration due to a contribution of resuspended topsoil particles.

The presence of polluting particles in the atmosphere can contribute substantial at the increased levels of trace elements in samples. The absorption and storage capacity of metal pollutants in the moss depend on the physicochemical properties and sorption capacity of the cell walls [25]. This second sampling was realized in summer (August); the mosses were exposed to different climatic conditions because during the period from May to October this region is characterized by the presence of rain and drastic temperature changes. Many elements may be partially released during precipitation in a solution, and prolonged precipitation may enable absorption of metals through the surface of the moss via active and passive processes. The rain may also have excessively washed the mosses and prevented the retention of pollutants in the moss structure [26]. In this case, it is possible that the increases of concentrations observed showed a flushing or leaching effect that resulted in decreased concentrations of contaminants in the moss structure [27].

3.2. Topsoil Analysis. The results of the elemental concentration in topsoils samples are presented in Table 3. The relative order of concentrations is for the four heavy metals in the topsoil samples, being $Cr > As > Hg >$ and Se , which demonstrated a different relationship when compared with results from the analysis of the mosses. Se concentrations were also significantly ($P < 0.05$) higher in topsoils than in mosses (Figure 2). The relative concentrations for the other elements were $Ce > Rb > Co > Sc > La > Sb > Cs > Eu >$

TABLE 2: Element concentration (mg kg^{-1}) in moss from the second sampling.

Sampling sites	As	Cr	Hg	Se	Co	Cs	Sc	Sb	Rb	Ce	Eu	La	Yb
Negrete (UR)	1.72 ± 0.16	32.47 ± 2.05	0.22 ± 0.06	1.96 ± 0.43	3.27 ± 0.16	1.13 ± 0.12	3.75 ± 0.15	1.11 ± 0.20	23.88 ± 5.39	4.11 ± 0.43	0.38 ± 0.10	2.08 ± 0.07	0.093 ± 0.02
Alameda (UR)	1.91 ± 0.13	29.28 ± 1.55	0.34 ± 0.08	1.98 ± 0.37	3.28 ± 0.18	0.81 ± 0.09	3.62 ± 0.14	1.74 ± 0.29	22.03 ± 5.02	5.87 ± 0.73	0.31 ± 0.08	3.47 ± 0.12	0.159 ± 0.03
Reforma (UR)	1.60 ± 0.09	23.42 ± 1.41	0.36 ± 0.07	1.11 ± 0.21	1.89 ± 0.10	0.67 ± 0.08	2.51 ± 0.10	2.02 ± 0.36	16.33 ± 3.81	9.92 ± 0.88	0.26 ± 0.07	5.41 ± 0.19	0.259 ± 0.05
Hípico (UR)	1.50 ± 0.10	17.52 ± 3.24	0.14 ± 0.06	1.60 ± 0.30	1.83 ± 0.10	0.54 ± 0.07	2.37 ± 0.09	1.33 ± 0.22	18.34 ± 4.14	3.13 ± 0.33	0.21 ± 0.06	2.19 ± 0.08	0.108 ± 0.02
Pilita (UR)	2.24 ± 0.13	19.05 ± 1.02	0.27 ± 0.07	2.20 ± 0.40	2.36 ± 0.13	0.70 ± 0.08	2.98 ± 0.12	1.00 ± 0.17	15.25 ± 3.50	4.28 ± 0.46	0.27 ± 0.07	2.80 ± 0.10	0.153 ± 0.03
Santín (UR)	1.92 ± 0.14	28.67 ± 1.80	0.43 ± 0.10*	2.24 ± 0.49	2.75 ± 0.14	1.06 ± 0.12	3.59 ± 0.14	1.42 ± 0.26	28.08 ± 6.25	3.11 ± 0.36	0.32 ± 0.09	1.94 ± 0.07	0.083 ± 0.02
Tolloacán (UR)	2.29 ± 0.20	48.59 ± 3.02*	0.32 ± 0.07	2.89 ± 0.62*	3.17 ± 0.16	1.01 ± 0.11	3.19 ± 0.13	3.07 ± 0.55*	24.46 ± 5.48	4.26 ± 0.50	0.40 ± 0.11	2.45 ± 0.09	0.115 ± 0.02
Lomas (TR)	0.94 ± 0.08	14.00 ± 0.75	0.20 ± 0.06	1.05 ± 0.20	5.41 ± 0.31*	2.60 ± 0.44*	1.53 ± 0.06	0.78 ± 0.13	9.39 ± 2.22	2.45 ± 0.27	0.13 ± 0.04	1.64 ± 0.06	0.078 ± 0.01
Acazulco (TR)	1.12 ± 0.07	13.12 ± 0.80	0.13 ± 0.03	1.27 ± 0.24	2.82 ± 0.14	0.88 ± 0.09	2.99 ± 0.12	0.62 ± 0.11	32.89 ± 7.20	12.18 ± 1.06*	0.33 ± 0.09	6.07 ± 0.21*	0.382 ± 0.08*
Pedregal (TR)	1.09 ± 0.10	10.10 ± 0.68	0.30 ± 0.08	1.10 ± 0.31	1.64 ± 0.09	0.71 ± 0.11	1.44 ± 5.86	0.80 ± 0.11	32.62 ± 7.21	2.28 ± 0.30	0.40 ± 0.11	1.50 ± 0.06	0.072 ± 0.01
San Miguel (TR)	2.41 ± 0.17*	28.03 ± 1.86	0.27 ± 0.06	2.77 ± 0.72	5.00 ± 0.24	1.71 ± 0.26	5.19 ± 0.21*	1.27 ± 0.18	28.30 ± 6.33	7.78 ± 0.73	0.59 ± 0.16*	4.43 ± 0.16	0.242 ± 0.05
Ameyalco (TR)	0.97 ± 0.09	6.37 ± 0.45	0.12 ± 0.03	0.75 ± 0.23	1.16 ± 6.25	0.46 ± 0.07	1.22 ± 5.04	0.44 ± 0.07	15.75 ± 3.59	2.35 ± 0.25	0.13 ± 0.03	1.47 ± 0.05	0.078 ± 0.01
San Antonio (PA)	1.73 ± 0.12	23.08 ± 1.46	0.24 ± 0.04	1.46 ± 0.32	2.60 ± 0.13	1.27 ± 0.14	2.90 ± 0.11	1.02 ± 0.18	33.26 ± 7.35*	3.10 ± 0.30	0.27 ± 0.07	2.01 ± 0.07	0.094 ± 0.07
San Diego (PA)	1.23 ± 0.06	7.82 ± 0.49	0.34 ± 0.21	1.05 ± 0.19	2.14 ± 0.11	0.75 ± 0.08	1.74 ± 0.07	0.75 ± 0.14	22.93 ± 5.14	8.20 ± 0.70	0.17 ± 0.04	4.82 ± 0.17	0.222 ± 0.04
Cacalomaacán (PA)	1.14 ± 0.10	9.13 ± 0.63	0.21 ± 0.04	1.66 ± 0.51	2.02 ± 0.10	1.32 ± 0.20	2.04 ± 8.30	0.29 ± 0.04	22.13 ± 4.94	2.99 ± 0.29	0.21 ± 0.06	1.64 ± 0.06	0.093 ± 0.02
Ciervita (PA)	1.24 ± 0.14	9.26 ± 0.63	0.23 ± 0.05	1.74 ± 0.53	1.96 ± 0.10	0.91 ± 0.14	2.21 ± 0.09	0.27 ± 0.04	16.30 ± 3.64	4.29 ± 0.38	0.26 ± 0.07	2.16 ± 0.08	0.093 ± 0.021

* Higher concentration.

TABLE 3: Element concentration (mg kg⁻¹) in topsoil sampled.

Sampling sites	As	Cr	Hg	Se	Co	Cs	Sc	Sb	Rb	Ce	Eu	La	Yb
Negrete (UR)	3.95 ± 0.34*	57.43 ± 1.21	2.13 ± 0.13	0.38 ± 0.02	19.91 ± 0.49	2.21 ± 0.09	16.05 ± 0.22	0.54 ± 0.06	52.94 ± 4.22	46.31 ± 0.876	1.77 ± 0.05	11.41 ± 0.49	1.04 ± 0.04
Alameda (UR)	1.60 ± 0.25	54.47 ± 1.19	3.17 ± 0.18	0.46 ± 0.02	13.79 ± 0.25	2.81 ± 0.08	14.12 ± 0.19	2.05 ± 0.08	63.17 ± 4.69*	69.97 ± 8.22*	1.87 ± 0.06	15.94 ± 0.49	1.06 ± 0.05
Reforma (UR)	1.56 ± 0.29	94.04 ± 2.66	0.55 ± 0.09	0.57 ± 0.05	16.90 ± 1.59	2.38 ± 0.08	17.03 ± 0.24	1.55 ± 0.08	53.01 ± 3.69	51.94 ± 1.05	1.95 ± 0.04*	11.82 ± 0.48	1.12 ± 0.12
Hípico (UR)	1.7 ± 0.09	65.93 ± 1.41	1.99 ± 0.23	0.36 ± 0.02	19.21 ± 0.35	2.69 ± 0.09	17.17 ± 0.24	0.78 ± 0.10	50.85 ± 4.27	23.17 ± 1.17	1.86 ± 0.04	13.37 ± 0.36	1.19 ± 0.05
Pilita (UR)	1.31 ± 0.20	50.76 ± 1.09	2.87 ± 0.16	0.27 ± 0.03	15.38 ± 0.31	2.26 ± 0.10	14.26 ± 0.21	$L_D = 0.75$	45.29 ± 3.89	48.83 ± 1.13	1.77 ± 0.04	10.57 ± 0.31	0.75 ± 0.03
Santín (UR)	0.80 ± 0.22	160.05 ± 4.87*	2.82 ± 0.25	0.40 ± 0.02	7.46 ± 0.22	3.89 ± 0.13*	24.19 ± 0.34*	3.22 ± 0.10	62.36 ± 4.69	66.23 ± 6.89	0.67 ± 0.03	12.49 ± 0.42	1.15 ± 0.05
Tolloacán (UR)	1.69 ± 0.31	73.07 ± 3.57	2.43 ± 0.16	0.65 ± 0.07	15.98 ± 0.29	2.65 ± 0.09	13.11 ± 0.18	2.51 ± 0.10	55.88 ± 2.92	55.41 ± 1.11	1.66 ± 0.02	12.96 ± 0.44	1.15 ± 0.09
Lomas (TR)	1.85 ± 0.31	87.70 ± 1.88	1.30 ± 0.22	0.42 ± 0.02	24.51 ± 0.43*	2.81 ± 0.16	20.69 ± 0.29	1.02 ± 0.13	62.41 ± 4.91	32.11 ± 1.08	1.79 ± 0.04	17.01 ± 0.49	1.40 ± 0.05
Acazulco (TR)	2.01 ± 0.28	133.50 ± 3.38	2.68 ± 0.22	0.73 ± 0.05	19.58 ± 0.34	2.17 ± 0.10	14.30 ± 0.21	0.56 ± 0.14	22.19 ± 4.41	37.86 ± 1.45	1.52 ± 0.04	19.79 ± 0.44	1.56 ± 0.09
Pedregal (TR)	1.88 ± 0.30	53.84 ± 1.20	2.38 ± 0.17	0.50 ± 0.03	11.25 ± 0.20	2.22 ± 0.09	10.54 ± 0.15	0.9 ± 0.03	31.62 ± 3.74	30.58 ± 1.09	1.39 ± 0.03	15.02 ± 0.31	1.12 ± 0.08
San Miguel (TR)	2.35 ± 0.29	114.61 ± 2.81	3.37 ± 0.45	0.54 ± 0.03	16.02 ± 0.29	2.03 ± 0.07	11.28 ± 0.16	0.89 ± 0.06	26.21 ± 2.05	39.29 ± 1.13	1.31 ± 0.03	19.78 ± 0.89	1.68 ± 0.12
Ameyalco (TR)	3.46 ± 0.63	113.78 ± 2.61	2.90 ± 0.20	0.60 ± 0.06	16.52 ± 0.33	2.31 ± 0.12	13.61 ± 0.19	1.25 ± 0.15	29.96 ± 4.40	39.29 ± 1.13	1.62 ± 0.04	20.73 ± 0.45*	1.79 ± 0.09*
San Antonio (PA)	1.65 ± 0.33	155.03 ± 4.23	2.93 ± 0.16	0.54 ± 0.06	19.50 ± 0.35	1.41 ± 0.06	14.26 ± 0.21	0.38 ± 0.08	18.64 ± 1.72	31.95 ± 0.67	1.11 ± 0.06	14.95 ± 0.50	1.38 ± 0.08
San Diego (PA)	3.72 ± 0.70	66.05 ± 4.80	3.38 ± 0.44*	0.78 ± 0.04*	10.06 ± 0.75	2.31 ± 0.12	5.45 ± 0.11	4.86 ± 0.33*	22.69 ± 5.49	52.25 ± 1.75	0.66 ± 0.04	18.51 ± 0.40	1.64 ± 0.10
Cacalomaacán (PA)	1.96 ± 0.26	51.45 ± 1.01	1.56 ± 0.21	0.42 ± 0.02	19.08 ± 0.34	2.99 ± 0.10	17.01 ± 0.24	0.52 ± 0.09	35.44 ± 3.37	33.68 ± 1.19	1.94 ± 0.04	17.03 ± 0.48	1.26 ± 0.04
Ciervita (PA)	1.59 ± 0.20	48.45 ± 0.88	1.21 ± 0.22	0.36 ± 0.02	16.23 ± 0.30	2.93 ± 0.11	14.24 ± 0.19	0.55 ± 0.09	34.93 ± 3.45	26.35 ± 1.11	1.67 ± 0.04	13.31 ± 0.39	1.06 ± 0.05

* Higher concentration.

FIGURE 2: Continued.

FIGURE 2: Element concentrations (mg kg^{-1}) in moss and soil samples. M1 (moss autumn), M2 (moss summer), and S: soil.

and Yb, whereas the sites located in Negrete (UR), San Diego (PA), and Ameyalco (TR) present the highest concentrations of Cr (Santín (UR), San Antonio (PA), and Acapulco (TR)); of Hg (San Diego (PA), San Miguel (TR), and Alameda (UR)); of Se (San Diego (PA), Acapulco (TR), and Tolloacán (UR)); of Co (Lomas (TR), Negrete (UR), and Acapulco

(TR)); of Cs (Santín (UR), Cacalomacán (PA), and Ciervita (PA)); of Sc (Santín (UR), Lomas (TR), and Hípico (UR)); of Sb (San Diego (PA), Santín (UR), and Tolloacán (UR)); of Rb (Alameda (UR), Lomas (TR), and Santín (UR)); of Ce (Alameda (UR), Santín (UR), and Tolloacán (UR)); of Eu (Reforma (UR), Cacalomacán (PA), and Alameda (UR)); of

La (Ameyalco (TR), San Miguel (TR), and Acazulco (TR)); of Yb (Ameyalco (TR), San Miguel (TR), and San Diego (PA)). Results of the soils analyses demonstrated that the highest average metal concentrations were registered in the total metal fraction. This result was expected by taking into account that this fraction includes elements of the organic fraction. The greatest differences were in levels of elements such as Cr, Hg, Co, Sc, Rb, Ce, and La. The measured concentrations demonstrated that the Ce concentration was greater than the La concentration, which may indicate that the presence of lanthanides and other elements in the topsoil samples was of natural origin [6].

The highest average As, Cr, Hg, and Se concentrations were found in samples from Negrete (for As), Santín (for Cr), and San Diego (for Hg and Se). The greatest differences between the results obtained for topsoils versus the mosses were observed in the concentrations of Cr, Cs, Co, Sc, Rb, Ce, Eu, La, and Yb. The elemental concentrations are variable by site (see Figure 2). The deposition of several of these elements could be attributed to anthropogenic activities, but some of the elements are found in the naturally occurring mineral matrix in topsoil samples. When the mosses in a predetermined area accumulate elements from the atmosphere, the soil in the area also acts as a receptor for these elements. This result was expected, as the measured samples included an organic fraction that was formed during the degradation and decomposition of plant tissues, and the elements contained in the plants pass into the topsoil. The deposition of contaminating particulates may have been increased by wind action, which enabled the transport and dispersion of pollution particles. The influence of other factors such as topsoil type, the presence of sedimentary rocks, the availability of nutrients and trace elements, geographical factors, altitude, topography, and wind direction (depending on the season) affected the concentrations of pollutants measured in the topsoil samples.

Unlike the pollutant concentration results obtained from mosses, it was shown that regions PA, TR, and UR do not follow a trend in pollution levels. In topsoils the highest concentrations of a large number of elements were obtained in samples from three areas (Figure 3). However, Cacalomacán and Ciervita, sites located in PA, showed lower levels of pollution in topsoil samples.

3.3. Statistical Analysis of As, Cr, Hg, and Se in Moss and Topsoil Samples. To define some possible correlations between the element content and pollution severity, PCA and CA were applied, based on the data of the average concentration values for As, Cr, Hg, and Se in the mosses from both of the sampling events (Tables 1 and 2).

For the first sampling (autumn) results data set 88.36%, the total variation of element variables is explained by two principal factors. Factor 1 (58%) could be related to elemental concentration and factor 2 (30.36%) related to industrial, natural source, and other life activities.

For PCA the results of the first sampling (autumn) showed that the sites (Figures 3(a), 3(c), and 3(e)) were segregated into three regions according to the degree of pollution: A (Reforma and Tolloacán (UR)), B (Ciervita and

Cacalomacán (PA) and Lomas (TR)), and C (Santín, Hípico, Negrete, and Alameda (UR)). Region A contained sites with a greater accumulation of As, Cr, Hg, and Se because they were more exposed to pollution sources, which increases their accumulation in mosses. The sites with the lowest pollution levels for the group of variables analyzed were located in Region B. The sites grouped in Region C had similar pollution levels that may have been influenced by similar pollution sources and their close proximity within the MAVT. San Antonio (PA) and Pilita (UR) were not included in any of the regions, indicating that these sites demonstrated distinct environmental characteristics uncommon to groups A (UR), B (PA, TR), and C (UR).

CA was applied to the data set as described above using Euclidian distances. The results are displayed as a dendrogram analysis (Figures 3(b), 3(d), and 3(f)). Three clusters were evident after analysis: cluster 1 (Tolloacán and Reforma (UR)), cluster 2 (Ciervita and Cacalomacán (PA) and Lomas (TR)), and cluster 3 (Santín, Alameda, Hípico, and Negrete (UR)). Observations for Pilita (UR) and San Antonio (PA) were not integrated to form a cluster, which could be indicators that the sites expressed distinct environmental characteristics in comparison with the others. The results of CA confirmed the results obtained with PCA.

The PCA results obtained in the second sampling (summer) (Figure 3(c)) showed three distinct regions: A (San Diego (PA) and Pedregal (TR)), B (Cacalomacán and Ciervita (PA)), and C (San Miguel (TR), Negrete, Tolloacán, and Pilita (UR), San Antonio (PA), and Alameda, Santín, and Reforma (UR)). Ameyalco and Lomas (TR) were not grouped. The graphical distances between all of the sampling points are very small, indicating the presence of similar concentration values among the sites. The cluster classification of the concentration results obtained in the second sampling is presented in Figure 3(d); three clusters were formed: (1) (Ciervita and Cacalomacán (PA)), (2) (San Diego (PA) and Pedregal (TR)), and (3) (Tolloacán, Reforma, Pilita, Santín, Alameda, and Negrete (UR), San Antonio (PA), and San Miguel (TR)). Ameyalco and Lomas (TR) were not included in the clusters, and this indicates that the concentrations at these sites were not similar to the other sites and confirmed the results obtained with PCA.

For the second sampling results data set (summer), 90.96% of total variation of element variables is explained by two principal factors. Factor 1 (73.96%) could be related to element concentration and factor 2 (17.00%) related to industrial, natural source, and other life activities.

The PCA of the topsoil sample measurements (Figure 3(e)) resulted in three regions: (a) (Pedregal (TR) and Alameda (UR)), (b) (Ciervita and Cacalomacán (PA)), and (c) (San Antonio (PA) and Ameyalco, San Miguel, and Acazulco (TR)). Samples derived from San Diego (PA), Tolloacán, Pilita Negrete, and Santín (UR), and Lomas and Reforma (TR) demonstrated a distant relationship to the other three regions with PCA, indicating that these sites have different environmental characteristics and exposure sources.

The CA from the topsoil samples was ambiguous because there was no clear relationship between the different sampling

FIGURE 3: Analysis of the elements (Se, Hg, Cr, and As). Mosses samples (autumn): (a) and (b); mosses samples (summer): (c) and (d); topsoil samples: (e) and (f).

sites. Each site exhibited different characteristics, and it can be assumed that the topsoil type at each site significantly influenced the composition of the samples. The dendrogram presented in Figure 3(f) illustrates the formation of three

clusters: (1) (San Miguel and Ameyalco (TR), San Antonio (PA), and Acapulco (TR)), (2) (Ciervita and Cacalomacán (PA)), and (3) (Alameda (UR) and Pedregal (TR)). Sites within each cluster are likely subjected to similar

environmental conditions. Sample results from Reforma, Pilita, Negrete, and Santín (UR), San Diego (PA), and Lomas (TR) were not included in any of the clusters, indicating significant environmental differences between these sites and those in the 3 clusters. This confirmed the results obtained with PCA.

For the topsoil sampling results data set, 70.58% of the total variation of element variables is explained by two principal factors. Factor 1 (46.24%) could be related to element concentration and factor 2 (24.34%) related to industrial, natural source, and other life activities.

4. Conclusion

Although mosses have a relatively long history of use as biomonitors, research on the uptake and localization of elements and on their sources has recently often been neglected. In this study, we evaluated the utility of mosses as biomonitoring tools to determine the elemental pollution levels in the air of MAVT as measured by INAA. The present result and literature data show that in remote and barren areas the elemental composition of unwashed moss is affected by entrapped crustal materials. The highest pollution levels were found in samples from Tollocán, Alameda, Reforma, and Santín (UR) and San Miguel (TR), which are all located in urban regions. The presence of high pollution levels in samples can be attributed to proximity of industrial areas that include a large number of vehicles, wind dynamics, and anthropogenic activities that contribute to the degradation of air quality. Samples from the site located at Cacalomacán (PA) contained the lowest levels of contamination; this is possibly because these regions are located far from industrial corridors with traffic flow and have limited public transport and low population density. This pattern of pollution could not be discerned with precision using the topsoil samples, as the highest concentrations of a large number of elements were distributed in these three areas. The results showed that studied moss species have a capacity to capture and/or incorporate elements present in the atmosphere at trace concentrations, making them a useful tool for use in environmental biomonitoring to assess air quality.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

Acknowledgments

The authors acknowledge Juan Vidal Molina and Sabino Hernández for their technical assistance. R. Mejía-Cuero acknowledges COMECYT for the M.S. fellowship received in support of this work. The authors acknowledge the support provided by the National Council of Science and Technology (CONACYT) and the State of Mexico Council of Science and Technology (COMECYT) (Grant no. EDOMEX-2009-C02-132003).

References

- [1] E. Vega, G. Sánchez-Reyna, V. Mora-Perdomo et al., "Air quality assessment in a highly industrialized area of Mexico: concentrations and sources of volatile organic compounds," *Fuel*, vol. 90, no. 12, pp. 3509–3520, 2011.
- [2] M. Aničić, M. Tomašević, M. Tasić et al., "Monitoring of trace element atmospheric deposition using dry and wet moss bags: accumulation capacity versus exposure time," *Journal of Hazardous Materials*, vol. 171, no. 1–3, pp. 182–188, 2009.
- [3] Y. V. Aleksiayenak, M. V. Frontasyeva, M. Florek et al., "Distributions of ^{137}Cs and ^{210}Pb in moss collected from Belarus and Slovakia," *Journal of Environmental Radioactivity*, vol. 117, pp. 19–24, 2013.
- [4] G. Vuković, M. A. Urošević, I. Razumenić et al., "Active moss biomonitoring of small-scale spatial distribution of airborne major and trace elements in the Belgrade urban area," *Environmental Science and Pollution Research*, vol. 20, no. 8, pp. 5461–5470, 2013.
- [5] C. Delgadillo and A. Cárdenas, *Manual de Briofitas*, Instituto de Biología, UNAM, 1990.
- [6] S. Dołęgowska and Z. M. Migaszwski, "Anomalous concentrations of rare earth elements in the moss-soil system from south-central Poland," *Environmental Pollution*, vol. 178, pp. 33–40, 2013.
- [7] S. Stankovic, P. Kalaba, and A. R. Stankovic, "Biota as toxic metal indicators," *Environmental Chemistry Letters*, vol. 12, no. 1, pp. 63–84, 2014.
- [8] A. Kłos, M. Czora, M. Rajfur, and M. Waclawek, "Mechanisms for translocation of heavy metals from soil to epigeal mosses," *Water, Air, and Soil Pollution*, vol. 223, no. 4, pp. 1829–1836, 2012.
- [9] B. Wolterbeek, S. Sarmiento, and T. Verburg, "Is there a future for biomonitoring of elemental air pollution? A review focused on a larger-scaled health-related (epidemiological) context," *Journal of Radioanalytical and Nuclear Chemistry*, vol. 286, no. 1, pp. 195–210, 2010.
- [10] V. Spagnuolo, S. Terracciano, and S. Giordano, "Trace element content and molecular biodiversity in the epiphytic moss *Lepidotodon smithii*: two independent tracers of human disturbance," *Chemosphere*, vol. 74, no. 9, pp. 1158–1164, 2009.
- [11] A. Travesi, *Análisis por activación neutrónica*, Servicio de Publicaciones de La J.E.N., Madrid, Spain, 3rd edition, 1975.
- [12] *Hypermet-PC, Version 3.0: For Automatic Analysis of Gamma-Ray Spectra*, Department of Nuclear Physics Institute of Isotopes, Budapest, Hungary, 1995.
- [13] M. V. Frontasyeva and S. S. Pavlov, "Analytical investigations at the IBR-2 reactor in Dubna," JINR Preprint E14-2000-177, Joint Institute for Nuclear Research, Dubna, Russia, 2000.
- [14] Statistica (data analysis software system), version 7, Statsoft, 2004, <http://www.statsoft.com/>.
- [15] L. M. Camacho, M. Gutiérrez, M. T. Alarcón-Herrera, M. D. L. Villalba, and S. Deng, "Occurrence and treatment of arsenic in groundwater and soil in northern Mexico and southwestern USA," *Chemosphere*, vol. 83, no. 3, pp. 211–225, 2011.
- [16] T. Ohnuki, F. Sakamoto, N. Kozai et al., "Application of the micro-PIXE technique for analyzing arsenic in biomat and lower plants of lichen and mosses around an arsenic mine site, at Gunma, Japan," *Nuclear Instruments and Methods in Physics Research, Section B*, vol. 190, no. 1–4, pp. 477–481, 2002.
- [17] Z. M. Migaszwski, A. Gałuszka, S. Dołęgowska, J. G. Crock, and P. J. Lamothe, "Mercury in mosses *Hylocomium splendens*

- (Hedw.) B.S.G. and *Pleurozium schreberi* (Brid.) Mitt. from Poland and Alaska: understanding the origin of pollution sources,” *Ecotoxicology and Environmental Safety*, vol. 73, no. 6, pp. 1345–1351, 2010.
- [18] F. Qarri, P. Lazo, T. Stafilov et al., “Multi-elements atmospheric deposition study in Albania,” *Environmental Science and Pollution Research*, vol. 21, no. 4, pp. 2506–2518, 2014.
- [19] M. Krachler, J. Zheng, R. Koerner, C. Zdanowicz, D. Fisher, and W. Shotyk, “Increasing atmospheric antimony contamination in the northern hemisphere: snow and ice evidence from Devon Island, Arctic Canada,” *Journal of Environmental Monitoring*, vol. 7, no. 12, pp. 1169–1176, 2005.
- [20] K. Isaac-Olivé, C. Solís, M. A. Martínez-Carrillo et al., “*Tillandsia usneoides* L, a biomonitor in the determination of Ce, La and Sm by neutron activation analysis in an industrial corridor in Central Mexico,” *Applied Radiation and Isotopes*, vol. 70, no. 4, pp. 589–594, 2012.
- [21] G. Zarazúa-Ortega, J. Poblano-Bata, S. Tejeda-Vega et al., “Assessment of spatial variability of heavy metals in metropolitan zone of toluca valley, Mexico, using the biomonitoring technique in mosses and TXRF analysis,” *The Scientific World Journal*, vol. 2013, Article ID 426492, 7 pages, 2013.
- [22] B. Caballero-Segura, P. Ávila-Pérez, C. E. Barrera Díaz et al., “Metal content in mosses from the Metropolitan Area of the Toluca Valley: a comparative study between inductively coupled plasma optical emission spectrometry (ICP-OES) and total reflection X-ray fluorescence spectrometry (TXRF),” *International Journal of Environmental Analytical Chemistry*, vol. 94, no. 13, pp. 1288–1301, 2014.
- [23] A. Cruzado, A. López, F. P. Escamilla et al., *Aire limpio: Programa para el Valle de Toluca 2007–2011*, Gobierno del Estado de México, Secretaría del Medio Ambiente, 2007.
- [24] R. Gerdol, L. Bragazza, and R. Marchesini, “Element concentrations in the forest moss *Hylocomium splendens*: variation associated with altitude, net primary production and soil chemistry,” *Environmental Pollution*, vol. 116, no. 1, pp. 129–135, 2002.
- [25] R. E. Andrus, J. M. Glime, B. H. Allen, and R. Ochrya, “Economic and ethnic uses of bryophytes,” in *Flora of North America North of Mexico*, Flora of North America Editorial Committee, Ed., vol. 27, pp. 14–41, 2007.
- [26] M. Tomašević, M. Aničić, L. Jovanović, A. Perić-Grujić, and M. Ristić, “Deciduous tree leaves in trace elements biomonitoring: a contribution to methodology,” *Ecological Indicators*, vol. 11, no. 6, pp. 1689–1695, 2011.
- [27] J. Poikolainen, E. Kubin, J. Piispanen, and J. Karhu, “Atmospheric heavy metal deposition in Finland during 1985–2000 using mosses as bioindicators,” *Science of the Total Environment*, vol. 318, no. 1–3, pp. 171–185, 2004.