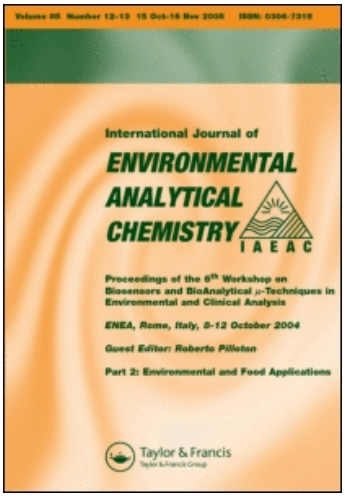


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# Atmospheric deposition of metals in romania studied by biomonitoring using the epiphytic moss *hypnum cupressiforme*

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# ATMOSPHERIC DEPOSITION OF METALS IN ROMANIA STUDIED BY BIOMONITORING USING THE EPIPHYTIC MOSS *HYPNUM CUPRESSIFORME*

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The results obtained from a study using the epiphytic moss species *Hypnum cupressiforme* to estimate heavy-metal atmospheric deposition in the province of Moldavia (Romania) are presented. The present study is a part of a biomonitoring programme in Moldavia and could serve as a starting point for further planning of biomonitoring using new moss species in regions where the commonly used *Hylocomium splendens* and *Pleurozium schreberi* are not present. Moss samples were collected within the catchment of Prut, a border river between Romania, Republic of Moldova and Ukraine, at a total of 25 sampling points and analysed for a wide range of elements, most of them heavy metals. The samples were analysed by instrumental neutron activation analysis using epithermal neutrons (ENAA) for 40 elements. Using inductively coupled plasma-mass spectrometry (ICP-MS), the elements Cd, Tl, Pb, V, Cr, Ni, Zn, As, Mo, Ag, In, and Bi were determined. Atomic absorption spectrometry (AAS) was used for the determination of Cd, Cu, Pb and Zn. Distribution maps were drawn for nine elements (As, Cd, Cr, Cu, Fe, Pb, Se, V, and Zn). By means of moss monitoring, it was possible to characterize most of the industrial and local sources of pollution within the studied area. The values are lower than those found in the western part of Romania, and the distribution of certain elements (As, Cu, Zn) reflects the predominant agricultural activities in eastern Romania. The highest levels of elements were found in the most industrialised and populated areas of Moldavia. Significant correlations were observed for the results obtained by different analytical techniques.

*Keywords:* Biomonitoring; Heavy metals; Mosses; ICP-MS; NAA; AAS

## INTRODUCTION

Among the compounds of anthropogenic origin, heavy metals emitted to the atmosphere in the form of solids or gases enter all ecosystems and, depending on their concentrations, could cause serious problems for plants, animals, and human beings. The conventional methods for measurement of atmospheric deposition of heavy metals using bulk precipitation samplers depend on special devices, technical installation,

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and frequent collection of samples, and are therefore restricted to a small number of sites within a larger area of investigation [1–3]. In recent decades, new reliable methods have been introduced that are simpler and cheaper than the traditional methods. Among these methods, the use of mosses and lichens for environmental monitoring has been systematically extended.

Mosses do not have a well-developed root system, and therefore they obtain their mineral nutrients mainly from precipitation and dry deposition of airborne particulates. Considering also that sampling and analysis are easier than by conventional methods, mosses have received increasing attention as suitable organisms for monitoring atmospheric deposition.

First introduced about 30 years ago [4,5], the use of mosses as biomonitors of heavy-metal atmospheric deposition is now a well-established technique [6–13]. Romania provided the first data on several moss-monitored metals between 1990 and 1992 [14]. In 1995, the moss technique was carried out to monitor atmospheric heavy-metal deposition in the eastern Romanian Carpathians and Transylvania [15,16]. This was the first time moss species had been used to monitor heavy-metal deposition in eastern Romania (province of Moldavia). When evaluating the suitability of the moss-biomonitoring technique for estimating atmospheric heavy-metal deposition, in eastern Romania localization of the sampling sites was difficult. In Romania, the common epigeic mosses used in biomonitoring studies occur in mountainous areas [15] but are rare in lowland places in eastern Romania and in the climatically dry regions in south-eastern Romania. Under such conditions, epiphytic moss species were considered as an alternative. In the eastern part of Romania (province of Moldavia), the most frequently found moss species is the epiphytic *Hypnum cupressiforme*. So far, very little is known about the feasibility of using epiphytic mosses in monitoring airborne heavy metals, although these mosses are used in areas where epigeic mosses are not present. There is an extensive literature presenting results of monitoring heavy-metal deposition by means of epiphytic mosses and comparing with results obtained using epigeic mosses but not referring details about the accumulation and retention processes and the factors influencing them. The literature contains very few references in terms of the influence of the phorophyte on the heavy-metal content of epiphytic mosses. Rasmussen and Johnsen [17] concluded that the metal content of the epiphytic moss *H. cupressiforme* originates mainly from the atmosphere, and in spite of the interspecific differences in bark composition, the same differences were not found in the concentrations of the mosses from different phorophyte species. Rasmussen [18] and Farmer *et al.* [19] concluded that the chemical content of epiphytic mosses growing on trees might reflect (to a limited extent) the chemistry of the tree bark.

The aim of this study was to present the regional atmospheric deposition of heavy metals in the province of Moldavia; to compare the results obtained by different analytical techniques; to compare the obtained results with data from other Romanian regions and other European countries; and to create a database for future surveys.

## EXPERIMENTAL

### Sampling

Samples of the epiphytic mosses *H. cupressiforme* were collected in Moldavia, within the catchment of the Prut River, at a total of 25 sampling sites (Fig. 1).

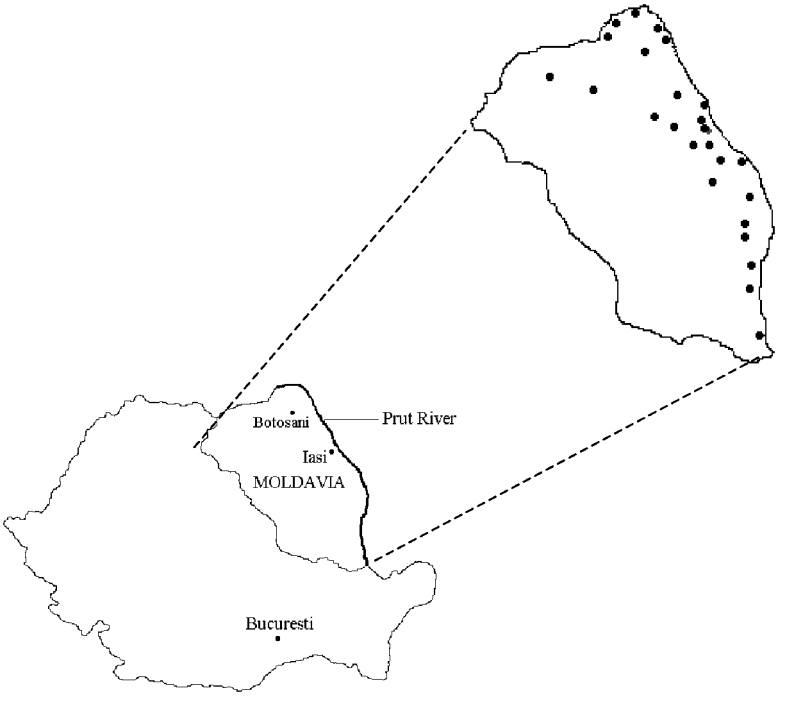


FIGURE 1 Sampling sites.



Sampling was carried out in 2000 (September–November) and 2001 (August). The sampling sites were located at least 300 m from main roads and populated areas, and at least 200 m from any road and single house. On each sampling site, four to six sub-samples were taken within an area of $50 \times 50$ m. Samples of the moss growing at a height of 0.5–1.5 m on deciduous trees (*Quercus* and *Fagus* species) were collected using disposable gloves (one pair for each sample), a spatula, and tweezers, all made from plastics, and stored in the field in polyethylene bags. In the laboratory, the samples were dried to constant weight at room temperature, and foreign matter was carefully removed. Only the green and greenish brown parts of the plants were used in the analysis. Although the age of the moss species could not be defined exactly, we assumed that the analysed material represents the average deposition for the last two years [12,20,21].

## Analysis

For the AAS and ICP-MS analysis, portions of about 0.4 g of moss were decomposed with 4 mL of concentrated nitric acid in a microwave oven. The extracts were then filtered, and demineralised water was added to a total volume of 25 mL. The presence of possible contaminants during the digestion process was controlled using blanks, one for every 11 samples. Accuracy was checked by analysis of three replicates of reference material M1, a *Pleurozium schreberi* moss reference sample prepared for calibration of laboratories participating in the 1995 European survey (supplied by the Finnish Forest Research Institute) [22]. The results are presented in Table I. Replicates were analysed for 50% of the samples. The Cu and Zn concentrations in the extracts were determined

TABLE I Determined and recommended [22] concentrations (mg/kg) in reference material M1

| *Element* | *Recommended* | *AAS* | *ICP-MS* |
|---|---|---|---|
| V | 0.60 ± 0.10 | – | 0.58 ± 0.01 |
| Cr | 0.50 ± 0.13 | – | 0.32 ± 0.03 |
| Ni | 0.99 ± 0.10 | – | 1.17 ± 0.05 |
| Cu | 3.18 ± 0.12 | 3.25 ± 0.01 | 3.5 ± 0.13 |
| Zn | 25 ± 0.9 | 24.1 ± 0.1 | 28.3 ± 0.6 |
| As | 0.070 ± 0.005 | – | 0.10 ± 0.004 |
| Mo | 0.08 ± 0.04 | – | 0.05 ± 0.007 |
| Ag | 0.024 ± 0.006 | – | 0.019 ± 0.001 |
| Cd | 0.077 ± 0.006 | 0.082 ± 0.005 | 0.10 ± 0.003 |
| In | 0.001 ± 0.000 | – | 0.0015 ± 0.0005 |
| Tl | 0.016 ± 0.002 | – | 0.014 ± 0.001 |
| Pb | 2.00 ± 0.13 | 2.64 ± 0.05 | 2.3 ± 0.09 |

by flame AAS (Perkin Elmer 1100). The concentrations of Cd and Pb were determined by graphite furnace AAS (Perkin Elmer AAnalyst 660).

V, Cr, Ni, Cu, Zn, As, Mo, Ag, Cd, In, Tl, Pb, and Bi were determined by high-resolution ICP-MS (Finnigan Element). Neutron activation analysis was performed at the Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, Dubna, Russia. Most elements were determined by ENAA. Samples of about 300 mg were irradiated in the IBR-2 fast pulsed reactor with a flux of about $10^{12}$ n/(cm$^2$ s) [23]. Two modes of analysis were employed: long irradiation by epithermal neutron activation analysis for 80 h in Channel 1 was used to determine elements associated with long-lived radionuclides (Ag, As, Au, Ba, Br, Ce, Co, Cr, Cs, Eu, Fe, Hf, K, La, Mo, Na, Ni, Rb, Sb, Sc, Se, Sr, Sm, Ta, Tb, Th, Zn, Zr, W, Yb, U) in moss samples packed in aluminium cups, whereas short irradiation of samples heat-sealed in polyethene foil for 3 min in Channel 2 was used by conventional NAA for short-lived radionuclides (Al, Ca, Cl, Cu, I, Mg, Mn, Ti, V). Gamma-ray spectra were recorded four times using a high-purity Ge detector; after decay periods of 5 min and 10 min following the short irradiation, and after 7 days and 17 days following the long irradiation. The accuracy was checked by analysis of certified reference material IAEA L-336 (Table II).



## RESULTS AND DISCUSSION

The results obtained for 45 elements are summarised in Table III. The values for the Moldavian area are clearly lower than corresponding levels in Transylvania and other areas in Romania [15,16] but comparable with those found in some other European countries [14,20,24]. It should be emphasized that, so far, the feasibility of epiphytic mosses with respect to monitoring of airborne heavy metals is not clearly known, and so any comparison with the results obtained using epigeic mosses should be made with caution.

In order to examine the dispersion of the elements and to identify the principal contaminated areas by these elements, maps of their distributions were drawn (Fig. 2). In the following, some of the determined elements are discussed with respect to spatial trends and contributing sources.

TABLE II Determined and certified concentrations (mg/kg) in reference material IAEA L-336

| | *EV* (mg/kg) ± *error* (%) | *CV* (mg/kg) ± *error* (%) |
|---|---|---|
| Na[a] | 0.29 ± 8.0 | 0.32 ± 8.0 |
| Cl[a] | 1.93 ± 2.2 | 1.90 ± 0.5 |
| K[a] | 1.78 ± 8.3 | 1.84 ± 0.5 |
| Sc | 0.168 ± 8.5 | 0.17 ± 6.4 |
| V | 1.70 ± 15 | 1.47 ± 8.8 |
| Cr | 1.12 ± 19.5 | 1.06 ± 8.7 |
| Mn[a] | 0.070 ± 6.2 | 0.063 ± 4.6 |
| Fe[a] | 0.422 ± 8 | 0.430 ± 2.5 |
| Co | 0.293 ± 15 | 0.287 ± 7.6 |
| Zn | 29.5 ± 6.4 | 30.4 ± 4.5 |
| As | 0.610 ± 20 | 0.639 ± 10.5 |
| Se | 0.18 ± 28 | 0.22 ± 0.5 |
| Br | 13.2 ± 25 | 12.9 ± 25 |
| Rb | 1.67 ± 36 | 1.76 ± 9.7 |
| Sr | 8.6 ± 21 | 9.3 ± 0.5 |
| Sb | 0.070 ± 14 | 0.073 ± 9.2 |
| Ba | 5.9 ± 12 | 6.4 ± 8.3 |
| Cs | 0.118 ± 16 | 0.11 ± 12.6 |
| La | 0.62 ± 15 | 0.66 ± 5.9 |
| Ce | 1.35 ± 18 | 1.27 ± 14.4 |
| Sm | 0.113 ± 6.5 | 0.106 ± 5.5 |
| Th | 0.137 ± 3 | 0.14 ± 1 |

[a]Element content (g/kg). EV: experimental values; CV: certified values.



## Arsenic

This element is emitted to the atmosphere mainly from coal combustion and mining. Other emission sources are the use of arsenic-based pesticides and steel production. The highest concentrations of this element were found in the south-eastern part of Moldavia. The use of pesticides (copper arsenates and cupric acetoarsenites) may have contributed to these increased values of As. In the south of Moldavia, higher As values are found near steelworks.

## Cadmium

Cd concentrations are relatively higher than in rural areas of other European countries [14]. The map of Cd distribution shows increased concentrations in the Iasi (the main city in the region) area and in the northern part of Moldavia (Botosani county). The high concentrations found in these areas could be associated with the coal combustion in thermo-electrical power stations. In the Iasi area, the emission of this element probably arises from the metallurgical and engineering industry.

## Chromium

Cr is emitted via the iron and steel processing industry and by coal combustion as main sources. In southern Moldavia, there is a large steel-processing plant, which could explain the consistently elevated values (13 mg/kg). Soil dust can be an important source of chromium as well.

TABLE III Concentrations (mg/kg) of elements in the moss species *Hypnum cupressiforme* in 2000 (Moldavia, Romania)

| *Element* | *Mean* | *Median* | *Range* |
|---|---|---|---|
| Na[a] | 480 | 380 | 220–1200 |
| Mg[a] | 2020 | 1820 | 980–3570 |
| Al[a] | 3290 | 2610 | 1110–8340 |
| Cl[a] | 300 | 270 | 180–510 |
| K[a] | 10 100 | 9520 | 7200–16 200 |
| Ca[a] | 7970 | 7670 | 4200–12 600 |
| Sc[a] | 0.52 | 0.36 | 0.16–1.58 |
| V[a] | 7.4 | 5.5 | 2.3–22.6 |
| Cr[a] | 6.2 | 4.3 | 1.3–19.1 |
| Mn[a] | 180 | 150 | 65–410 |
| Fe[a] | 2020 | 1370 | 620–5760 |
| Co[a] | 0.61 | 0.52 | 0.21–1.77 |
| Ni[a] | 4.2 | 2.9 | 1.7–11.3 |
| Cu[b] | 10.6 | 10.0 | 5.0–19.9 |
| Zn[b] | 36 | 32 | 20–86 |
| As[a] | 0.78 | 0.66 | 0.27–1.93 |
| Se[a] | 0.17 | 0.16 | 0.03–0.43 |
| Br[a] | 5.8 | 6.1 | 1.9–11.6 |
| Rb[a] | 9.5 | 7.9 | 3.5–24.9 |
| Sr[a] | 39 | 35 | 17–92 |
| Zr[a] | 69.6 | 54.9 | 23.4–207 |
| Mo[a] | 0.66 | 0.62 | 0.19–1.3 |
| Ag[a] | 0.058 | 0.068 | 0.015–0.095 |
| Cd[b] | 0.44 | 0.41 | 0.25–1.01 |
| In[c] | 0.0079 | 0.0072 | 0.0031–0.0163 |
| Sb[a] | 0.175 | 0.162 | 0.086–0.385 |
| I[a] | 1.96 | 1.94 | 0.82–3.48 |
| Cs[a] | 0.29 | 0.21 | 0.11–0.94 |
| Ba[a] | 78 | 88 | 35–124 |
| La[a] | 2.47 | 1.94 | 0.77–6.9 |
| Ce[a] | 9.7 | 6.8 | 2.4–29.4 |
| Sm[a] | 0.31 | 0.20 | 0.11–1.08 |
| Tb[a] | 0.048 | 0.036 | 0.027–0.132 |
| Yb[a] | 0.16 | 0.13 | 0.05–0.42 |
| Hf[a] | 0.53 | 0.43 | 0.19–1.62 |
| Ta[a] | 0.056 | 0.042 | 0.02–0.162 |
| W[a] | 0.18 | 0.15 | 0.07–0.43 |
| Au[a] | 0.0063 | 0.0037 | 0.0016–0.0361 |
| Tl[c] | 0.043 | 0.041 | 0.016–0.077 |
| Pb[b] | 14.0 | 11.2 | 7.1–32 |
| Bi[c] | 0.058 | 0.052 | 0.028–0.142 |
| Th[a] | 0.47 | 0.31 | 0.14–1.46 |
| U[a] | 0.16 | 0.12 | 0.05–0.53 |

[a]Determined by ENAA. [b]Determined by AAS. [c]Determined by ICP-MS.



## Copper

A number of local sources (e.g. the counties of Iasi and Vaslui) are responsible for the distribution of Cu, owing to the usage of copper-based fungicides, bactericides and fertilizers in vineyards (the aforementioned counties being big wine-producers).

## Iron

This element may be attributed to dry deposition of wind-blown soil particles and dust on the moss, considering the high correlation with aluminium, chromium and scandium

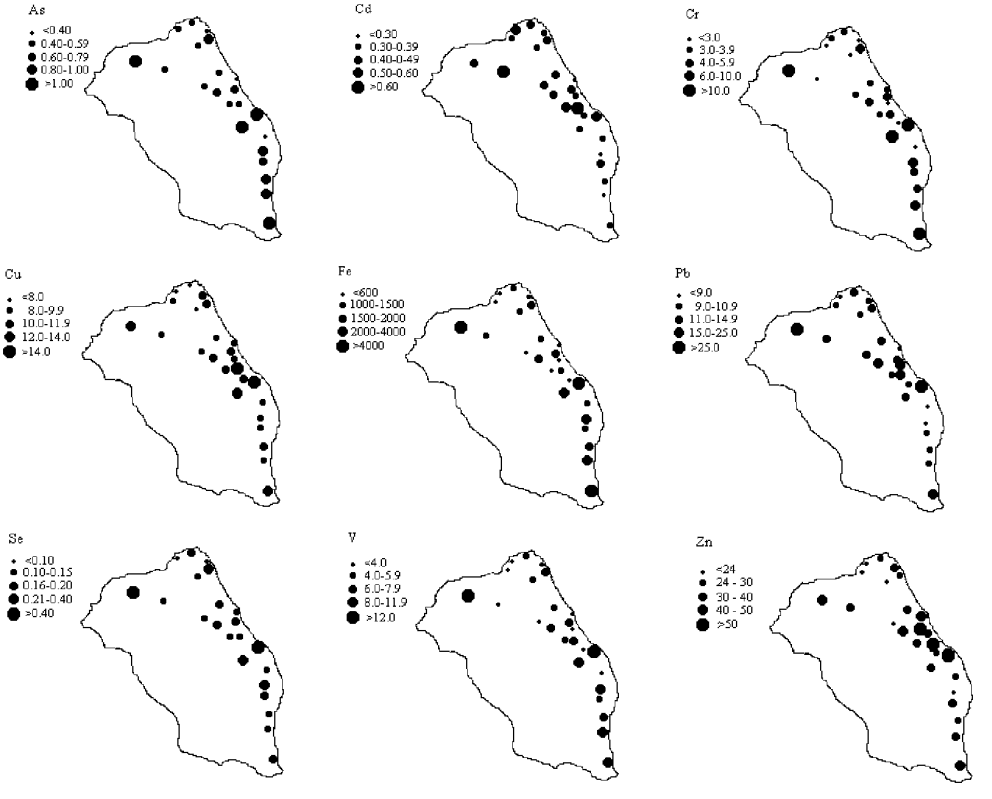


FIGURE 2 Spatial distribution of element concentrations (mg/kg) in sampling carried out in 2000.



(crustal elements). The highest concentrations (5700 mg/kg) in the southern regions may be caused by major emissions from the iron and steel industry.

## Lead

Lead is a local pollutant resulting from industries and from combustion of leaded fuel. In Romania, leaded gasoline has not been totally substituted by lead-free gasoline. Higher Pb concentrations have been found in the densely populated and industrialised parts of Moldavia (Iasi and Botosani). There seems to be a relationship between Pb patterns and traffic density, considering the tendency towards decreasing values for the 2001 samples in the densely populated areas compared with those for 2000 samples.

## Selenium

The distribution pattern of Se is homogeneous, thus possibly indicating contributions from other regions outside the studied area, such as Transylvania (western Romania), where the Se concentration has been found to be relatively high [16]. Selenium is typically found among pollutants transported over wider areas [24].

### Vanadium

A consistently higher level of this element compared with those in other European countries [20,25] is generally present in the studied area, probably due to contributions from Transylvania, where high concentrations have been obtained for this element [16]. Soil dust may be another source. Local concentration maxima of V have been observed in the southern regions (iron industry).

### Zinc

The relatively constant 'base' level of 20–40 mg/kg generally present in all regions may be explained by the contribution from tree canopies, considering that both moss species are epiphytic, growing on trees. The highest levels of Zn have been found in Iasi county, where there are metallurgical and engineering industries. There may also be contributions from Zn-based insecticides used for fruit trees and fungicides for fruits, as this area has many orchards.

For the elements determined in the samples collected in 2000 and 2001, the values are similar, indicating no great change in atmospheric deposition from one year to another (Table IV). Significant differences ($t$ test and ANOVA test) were observed only for Zn and Ag. This may suggest the existence of several constant pollution sources in the area studied.

Among the seven elements determined by both NAA and ICP-MS, significant correlations have been observed for V, As ($p < 0.001$), Cr, Ni, Zn ($p < 0.01$), and Mo ($p < 0.05$) (Table V). It should be noted that NAA determines the total content of elements in the samples, whereas ICP-MS determines only the fraction soluble in nitric acid. The values obtained by ICP-MS for the crustal elements are in good agreement with the results obtained by NAA on the same samples. This supports the following conclusions: the contamination of samples with soil particles is relatively low; the elemental content of the mosses is mainly from the atmosphere; the contamination during sampling was minimal; the accuracy of the analyses is good. These results encourage us to use complementary techniques in future

TABLE IV Ratios between element contents (determined by ICP-MS) in *Hypnum cupressiforme* in the 2001 and 2000 samplings

| *Element* | *Mean ratio ± SD* | *Significance* |
|---|---|---|
| Cd | 1.03 ± 0.28 | n.s.[a] |
| Tl | 0.96 ± 0.35 | n.s. |
| Pb | 0.90 ± 0.34 | n.s. |
| V | 0.87 ± 0.24 | n.s. |
| Cr | 0.89 ± 0.28 | n.s. |
| Ni | 0.85 ± 0.28 | n.s. |
| Cu | 1.00 ± 0.35 | n.s. |
| Zn | 1.33 ± 0.32 | * |
| As | 0.89 ± 0.22 | n.s. |
| Mo | 0.93 ± 0.28 | n.s. |
| Ag | 0.83 ± 0.36 | n.s. |
| In | 0.99 ± 0.37 | * |
| Bi | 0.95 ± 0.37 | n.s. |

[a]n.s.: not significant. *$p < 0.05$.

TABLE V Correlation coefficients between element concentrations in moss analysed by different methods

| *Element* | *Methods* | *R* | *Slope* | *Intercept* |
|---|---|---|---|---|
| V*** | | 0.97 | 1.64 | −1.33 |
| Cr** | | 0.92 | 2.59 | −1.76 |
| Ni** | | 0.66 | 1.19 | −0.26 |
| Zn** | ICP-MS/NAA | 0.75 | 0.84 | 7.19 |
| As*** | | 0.88 | 0.70 | −0.10 |
| Mo* | | 0.66 | 1.30 | 0.24 |
| Ag | | 0.12 | 0.22 | 0.05 |
| Cd** | | 0.75 | 0.85 | 0.02 |
| Pb*** | ICP-MS/AAS | 0.96 | 0.90 | 2.10 |
| Cu*** | | 0.94 | 0.73 | 1.82 |
| Zn*** | | 0.91 | 0.95 | 0.08 |

*$p < 0.05$; **$p < 0.01$; ***$p < 0.001$.

moss analyses. Table V shows the good correlation between the Cu, Pb and Zn values obtained by ICP-MS and AAS.



## CONCLUSION

By means of the moss biomonitoring technique, it was possible to characterize the spatial distribution of heavy metals in the investigated area. The values obtained are comparable with those found in other European countries but clearly lower than in the western part of Romania. No significant differences were observed between the 2000 and 2001 values, thus indicating no major changes in the atmospheric deposition of the pollutants under investigation between the two years. The values obtained by ICP-MS agree well with the results obtained by NAA on the same samples.

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