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Air Pollution Studies in Albania Using the Moss Biomonitoring Technique

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Abstract:

An exposure to heavy metals is a significant problem, not only because of environmental damage but also because it affects human health and represents a key economic problem. Thus, it seems very important to develop and improve a long-term passive monitoring technique to assess the type and level of heavy metal pollution of any particular areas. Mosses are useful indicators for biological monitoring of regional atmospheric depositions and heavy metal contamination of their environment. This study represents the first application of moss biomonitoring technique in Albania. Samples of terrestrial mosses Hypnum cupressiforme were collected at 13 sites during October-September 2010 following the the guidelines of the UNECE ICP Vegetation. The elements like aluminium, chromium, iron, nickel and vanadium were analysed by ICP-AES technique. Geographical distribution maps of the elements over the sampled territory were constructed using GIS technology. The most contaminated sites with heavy metals resulted those with high road traffic, high population

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density and in the vicinity of different residential and industrial activities.

Key words: air pollution, biomonitor, moss, heavy metals, ICP-AES.

Introduction

An exposure to heavy metals is a significant problem of environmental toxicology. Heavy metal contamination of the biosphere has increased sharply since 1900 (Nriagu, 1979). These metals even if deposited constantly in small rates over long periods of time, accumulate in the environment and will probably pose an increasing major environmental and human health hazard in future (Walkenhorst et al, 1993). Since heavy metals are elements, they cannot be disintegrated, so they will remain and accumulate in the environment. There are a multitude of anthropogenic emissions of heavy metals in the environment. Generally heavy metals enter into the environment mainly via three routes: (i) deposition of atmospheric particulates (e.g. mining, smelting, fossil fuel combustion, municipal waste incineration, cement production and phosphate mining). (ii) disposal of metal enriched sewage sludge and sewage effluents, commercial fertilizers and pesticides and animal waste specially to the terrestrial and aquatic environment, (iii) by-product from metal mining processes (Shrivastav, 2001; Smodis & Bleise, 2000). Among them, the major source of metals is from mining and smelting. Mining releases metals to the fluvial environment as tailings and to the atmosphere as metal-enriched dust whereas smelting releases metals to the atmosphere as a result of high-temperature refining processes (Adriano, 1986). Because of these elements do not decay with time, their emission to the environment is a serious problem which is increasing worldwide due to the rapid growth of population, increasing

combustion of fossil fuels and the expansion of industrial activities (Smodis & Bleise, 2000).

The effect of airborne materials on the quality of the human environment has prompted many investigators to search for means of measuring ambient atmospheric concentrations of these materials. In addition to the pollution levels recorded in densely populated and industrial areas, consideration must also be given to the dispersion of contaminants in the air and their transportation. Thus, it seems very important to develop and improve a long-term passive monitoring technique to assess the type and level of heavy metal pollution of any particular areas.

In many European countries increased efforts to establish heavy metal monitoring have led to a number of environmental programmes at the national and international levels. The moss technique introduced in Scandinavian countries about 25 years ago has shown to be most suitable for studying the deposition of heavy metals. It has found numerous applications and is now being widely used for large scale deposition studies (O. Berg *et al.*, 1995; E. Steinnes *et al.*, 1994). The bioaccumulation properties of mosses make them very efficient indicators for atmospheric metals and trace element depositions (Rühling and Tyler, 1968). The use of native terrestrial mosses as biomonitors is now a well-recognized technique in studies of atmospheric contamination (Fernandez and Carballeira, 2002) and is applied as a practical tool in establishing and characterizing deposition sources. For the first time this technique has been applied to a systematic study of air pollution with heavy metals. This study is only a part of the survey that covers all the territory of Albania. With this survey Albania attended for the first time the *European moss survey* in 2010/11, through the *UNECE ICP Vegetation Programme*. The aim of this study was the monitoring of air pollution at three regions in Albania, identifying the most polluted areas and defining different pollution sources.

Materials and Methods

Hypnum cupressiforme moss samples were collected at three regions of Albania (Tirane, Durres and Lezhe), following the guidelines of the UNECE ICP Vegetation. The moss samples were collected during September-October 2010 at 13 locations. In the laboratory, after manual removal of all adhering material (plant remains, soil particles, etc.), only the green–brown moss shoots were subjected to analysis as they correspond approximately to the deposition over the last three years. Then the moss samples were dried to constant weight at 30–40°C for 48 h and homogenized to a fine powder in agate mill.

Moss samples were digested by using of Microwave digestion system (Marsx, CEM, USA). All of the reagents used for this study were with analytical grade: nitric acid, trace pure (Merck, Germany), hydrogen peroxide, p.a. (Merck, Germany), and redistilled water. About 0.5 g of moss material was placed in a Teflon vessel and treated with 7 ml of concentrated nitric acid (HNO_3) and 2 ml hydrogen peroxide (H_2O_2) overnight. The procedure was continued with full digestion of moss material in microwave digestion system (Mars, CEM, USA). Digests were filtrated and quantitatively transferred to 25 ml calibrated flasks.

The content of elements in the moss samples were determined by inductively coupled plasma – atomic emission spectrometric (ICP-AES) method performed at the "*Institute of Chemistry, Faculty of Science, Sts. Cyril and Methodius University, Skopje, Macedonia*". The certified M2 and M3 moss samples were used for quality control of ICP-AES analysis. These certified moss samples were prepared within the monitoring program "The European Moss Survey".

Results and Discussions

In Figure 1. is shown the map of Albania and some neighbouring countries and also the map of the survey area together with the locations of sampling sites. The names of sampling sites of moss samples and the content of elements Al, Cr, Fe, V and Ni (in µg/g dry weight) are given in Table 1. In Figures 2-5 are shown the maps of geographical distribution of elements Al, Cr, Fe and V. These maps were prepared at "*Joint Institute for Nuclear Research, Dubna Moscow Region, Russia*", using the Geographic Information System (GIS).

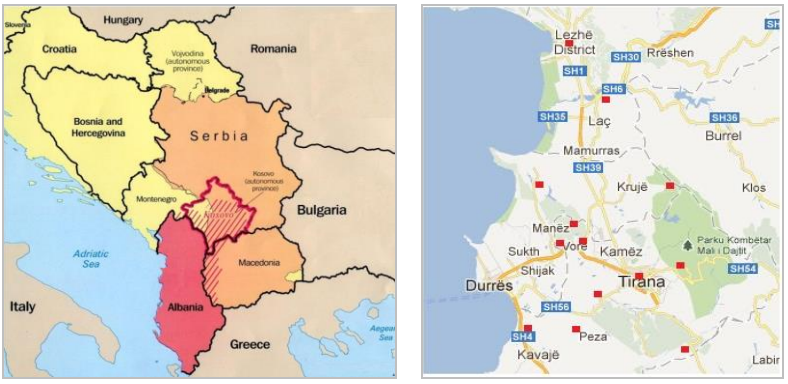

Figure 1. Republic of Albania (left) and survey area with the location of sampling sites (red dots-right).

Table 1. The sampling sites of moss samples and the content of elements obtained in this study (µg/g, dry weight).

Sampling sites	Elements				
	Al	Cr	Fe	V	Ni
Berxulle	806	2.58	642	1.68	2.21
Karrec	1520	4.26	1162	2.91	3.85
K. Botanik	1051	3.51	901	2.29	2.60
Dajt	573	2.40	469	1.15	3.55
Lalm	1269	4.42	1077	2.47	3.64
Ndroq	1267	5.15	1181	2.58	3.74
Shkafane	1625	4.36	1351	2.57	6.73
Manez	2254	5.76	1932	4.34	7.54
Golem	2331	11.68	2221	4.32	13.01
Krrabe	1244	3.34	1462	2.93	3.33
Milot	2722	7.00	3538	6.03	11.36

Lezhe	1712	6.12	2383	16.94	11.27
Kruje	1492	8.28	1694	5.00	5.29

Our results are compared with the corresponding data from similar studies in the neighboring Balkan countries (Frontasyeva et al., 2004; Spiric et al., 2009; Barandovski et al., 2008). In Table 2. are given the ranges (min-max) of elements. For comparison with a pristine territory, the corresponding data for northern Norway (E. Steinnes, 2007) are shown also in the right-hand column. As it is seen from Table 2. the maximum values of each element in our results are within the ranges reported from all other countries, except Norway. So, the results of our study show that in comparison to the Balkan countries our survey area is cleaner, however, compared to a clean area such as Norway our area is influenced by pollution.

Table 2. Comparison of the results obtained in the present study with other Balkan countries and Norway (µg/g, dry weight).

Elements	Albania	Serbia	Romania	Kroatia	Macedonia	Norway
Al	573-2722	1280-22090	830-23000	398-21460	825–17600	67-820
Cr	2.40-11.68	1.14-22	2.72-51.9	0.76-33	2.33-122	0.10-4.2
Fe	642-3538	720-9230	815-21340	320-12140	424-17380	77-1370
V	1.15-16.94	2.85-39	2.85-39	0.91-32	1.79-43	0.39-5.1
Ni	2.21-13.01	1.96-26	0.6-32	0.66-18	0.09-24	0.39-5.1

Aluminium

Aluminium is the third most abundant element in the lithosphere. Most naturally occurring aluminium compounds are sparingly soluble and bioavailable; therefore, relatively small quantities of aluminium are found in most biological samples unless contaminated with soil (dust) (Pais *et al.*, 1997). The largest source of airborne aluminium-containing particulates is the flux of dust from soil and the weathering of rocks (Lee and Von Lehmden 1973; Sorenson et al. 1974). Human activities, such as mining and agriculture, contribute to this wind-blown dust (Eisenreich 1980; Filipek et al. 1987). The major anthropogenic sources of aluminium-containing

particulate matter include coal combustion, aluminium production, and other industrial activities, such as smelting that process crustal minerals (Lee and Von Lehmden 1973). Away from local pollution sources, aluminium is a good indicator of mineral particles, mainly windblown soil dust, as it is present at high concentrations in the earth's crust. Therefore, the spatial pattern of aluminium concentrations in mosses might provide an indication of the contribution of wind re-suspension to the deposition of metals to mosses, reflecting to some extent historical deposition of heavy metals. A higher accumulation of soil dust does not necessarily translate into a higher deposition flux for all metals in the same way. The deposition flux of metals depends on the particle size distribution.

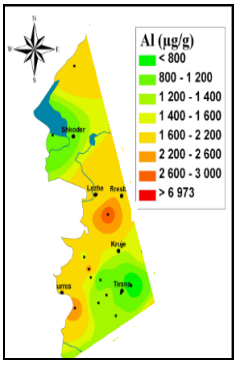

Figure 2. The geographical distribution of element Al.

The geographical distribution of aluminium is typical of the group of crustal elements predominantly supplied to the moss by windblown soil dust, showing relative uniform mean values between regions. Enhanced levels of aluminium in mosses are observed mainly at Milot (2722 µg/g), Golem (2331 µg/g) and Manez (2254 µg/g) and this seems to be related to the soil dust created during different human activities in these areas. In Golem it is related to the soil dust created as a result of diggings, because massive constructions have been and still continue to be carried out in this area. While in Milot and

Manez aluminium is related to the soil dust created from agricultural activities in these lands. Previously in Milot existed a place which was used for the collection of various minerals coming from different parts of Albania, in order to be transported later to different destinations by train, for example to the ex-chemical-metallurgical factory in Laç or other factories or to the port of Durres to be exported. So, the enhanced level of aluminium in Milot could also originate from the past activities related to the collection of minerals in this area and also as a result of processing various minerals at the ex-chemical-metallurgical factory in Laç (near Milot).

Chromium

Chromium is present in the Earth's crust, with the main natural source of exposure being continental dust present in the environment (Barnhart 1997; Fishbein 1981; Pellerin and Booker 2000). Chromium is released into the environment in larger amounts as a result of human activities, which account for 60–70% of the total emissions of atmospheric chromium (Alimonti et al. 2000; Barceloux 1999; Seigneur and Constantinous 1995), mainly by stationary point sources, including industrial, commercial, and residential fuel combustion, *via* the combustion of natural gas, oil and coal, chrome plating and steel production (Kimbrough et al. 1999; Pacyna and Pacyna 2001; Seigneur and Constantinous 1995; EPA 1990b). Approximately one-third of the atmospheric releases of chromium are believed to be in the hexavalent form (Johnson et al. 2006). Other potentially small sources of atmospheric chromium emission are cement-producing plants (cement contains chromium), the wearing down of asbestos brake linings that contain chromium, incineration of municipal refuse and sewage sludge, and emission from chromium-based automotive catalytic converters. Emissions from cooling towers that previously used chromate chemicals as rust inhibitors are

also atmospheric sources of chromium (EPA 1984b, 1990b; Fishbein 1981).

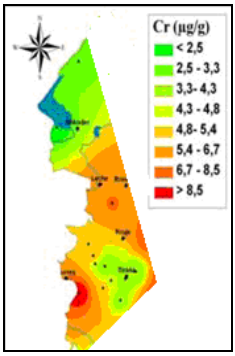

Figure 3. The geographical distribution of element Cr.

The highest value of chromium was found in the area of Golem (11.68 µg/g) and then in Kruje (8.28 µg/g) and Milot (7.00 µg/g). One source of chromium in Golem is related to the port of Durres (near Golem), where in an area of 90 thousand m^2 are deposited 360 thousand tones of ores like chromium and ferrochromium and also thousand tones of coal, which especially in windy weather raise black clouds of dust into the air. Another source may be the ex-chemical enterprise in Durres, which in the past used to produce potassium dichromate for leather tanning. In some areas, only a few kilometers around the enterprise, have been deposited in inappropriate conditions thousands of tones of toxic wastes, among which potassium dichromate and other wastes rich in chromium. Air pollution from soil dust created during processes of massive constructions which have been carried out in this area and around it, as well as from the cement used, may be other sources of pollution with chromium in Golem. In Kruje, chromium is found in the environment as a result of burning various combustible materials in lime kilns (which have been operating for several years in an uncontrolled manner in this area) or in the cement factory, as well as from the cement itself produced in this factory, activities that spread in the

environment clouds of smoke and dust. The content of chromium that is found in Milot has the origin from batches of chromium minerals that previously used to be placed in this area in order to be transported later to different destinations by train.

Iron

Iron is the fourth most common element in the Earth's crust and perhaps the main component of the depth of our planet. It is present in varying amounts in many rocks. Iron is released to the environment from both natural sources and by anthropogenic processes. The largest sources of airborne iron-containing particulates are the weathering of rocks and the flux of dust from soil especially in environments with rare vegetation and agricultural regions. The main anthropogenic emission sources of iron to the environment are mining, iron and steel industry which can discharge iron in the environment as emission from the process of melting and as dust from mineral crushing process, extraction and burning of coal and intensive traffic (Rühling and Steinnes, 1988).

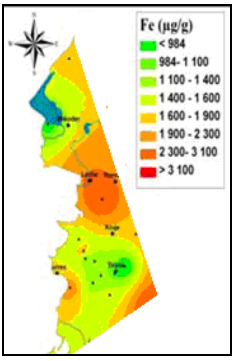

Figure 4. The geographical distribution of element Fe.

In Milot is found the highest level of iron (3538 µg/g), followed by Lezhe (2383 µg/g), Golem (2221 µg/g) and Manez (1932 µg/g). This elevated level of iron in Milot has its origin from the past activities of ex-chemical-metallurgical factory located in Laç

(near to Milot). In this factory, minerals like pyrite (FeS_2) and chalcopyrite ($CuFeS_2$) have been used for copper extraction as well as for sulphuric acid production which later was used for phosphate fertilizer production. Also, another source of iron in Milot are the batches of chromium minerals that previously used to be deposited in this area. In Golem the elevated level of iron may originate from the soil dust created during the processes of massive constructions and also from iron minerales like ferrochromium and iron-nickel deposited in the port of Durres. In Lezhe the high level of iron is related to windblown soil dust, hence to the geology of this area which is rich in titanomagnetite mineral. While in Manez the high content of iron is related to the dust created during agricultural activities in this area.

Vanadium

Vanadium is widely distributed in the earth's crust at an average concentration similar to that of zinc and nickel (Byerrum 1991). It is also found in phosphate rock and certain ores and is present in some crude oils as organic complexes (Lide 2008). Vanadium is released naturally to the atmosphere by the formation of continental dust, marine aerosols and volcanic emissions (Byerrum et al. 1974). The quantities entering the atmosphere from each of these sources are uncertain; however, continental dust is believed to account for the largest portion of naturally emitted atmospheric vanadium followed by marine aerosols (Zoller et al. 1973). Combustion of heavy fuels, especially in oil-fired power plants, refineries and industrial boilers, and coal are the major source of anthropogenic emissions of vanadium into the atmosphere (Mamane and Pirrone 1998; Sepe et al. 2003). Anthropogenic releases of vanadium to soil are less widespread than natural releases and occur on a smaller scale. These include the use of certain fertilizers containing materials with high vanadium content such as rock phosphate, superphosphate, and basic slag

as well as disposal of industrial wastes such as slag heaps and mine tailings. The form of vanadium present in the soil is determined largely by the parent rock. Ferric hydroxides and solid bitumens (organic) constitute the main carriers of vanadium in the sedimentation process. Iron acts as a carrier for trivalent vanadium due to the high affinity between trivalent vanadium and trivalent iron (Van Zinderen Bakker and Jaworski 1980).

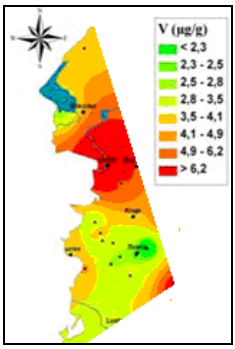

Figure 5. The geographical distribution of element V.

The highest level of vanadium is found in Lezhe (16.94 µg/g). This area contains titanomagnetite minerals which are known to be rich in vanadium, so this elevated concentration of vanadium is related to the geology of this area, thus with the windblown soil dust. The levels of vanadium in other areas are lower, however among them can be distinguished Milot (6.03 µg/g) and Kruje (5.00 µg/g) followed by Manez (4.34 µg/g) and Golem (4.32 µg/g). In Milot the vanadium content is related to the ex-chemical-metallurgical factory located in Laç, where considerable quantities of used vanadium catalyst are deposited inappropriately. In Kruje the origin of vanadium is from burning combustible materials in lime kilns that used to operate for a long time in this area, like coal or oil residues, and from burning combustible materials in the cement factory. In Manez vanadium can be present in environment as a result of the use of vanadium-containing fertilisers like superphosphates

in agriculture, while in Golem vanadium is found as a result of soil dust created during construction processes and as well as from marine aerosols.

Nickel

Nickel releases to the atmosphere occur from natural discharges such as windblown dust, volcanic eruptions vegetation, as well as from anthropogenic activities which exceed five times that from natural sources (Nriagu and Pacyna 1988) and they include the burning of residual and fuel oil, which is responsible for 62% of anthropogenic emissions, followed by nickel mining, smelting, refining, production of steel and other nickel-containing alloys, fossil fuel combustion, wood combustion, waste incineration and road traffic (Bennett 1984; Schmidt and Andren 1980).

Regarding to the content of nickel found in mosses, the areas of Golem (13.01 µg/g), Milot (11.36 µg/g) and Lezhe (11.27 µg/g) can be distinguished. In Golem, besides the windblown soil dust created from the processes of massive constructions, nickel comes also from the iron-nickel minerals deposited at the port of Durres. Also, all of these three monitoring areas have national roads in their vicinity, reflecting so the pollution from road traffic.

A Pearson′s correlation test was carried out to investigate the correlation between metal concentrations (significance was attributed to values of $P < 0.05$). Results have indicated highly positive significant correlations ($r > 0.5$) between the crustal element Al and the elements Fe, Cr, Ni, V, which indicates for their dual origin, geogenic and anthropogenic. Their geogenic origin is related to the deposition of windblown soil dust in mosses. The fact that all these elements have high levels in Golem, Milot and Manez, indicates that their origin is also from the soil dust created from various anthropogenic activities, like the windblown soil dust resulting from massive constructions, from the depositions of various

minerals and coal and from the agricultural activities. Other anthropogenic activities are the burning of various combustible materials and road traffic.

Conclusions

This was the first study of the atmospheric environment within the Republic of Albania and it adds this country to the European moss network. In comparison with neighboring countries where similar studies have been made, the results obtained for Albania, are within the ranges reported from all other countries, except Norway. So, the results of our study show that in comparison to the Balkan countries our survey area is cleaner, however, compared to a clean region such as Norway our area is influenced by pollution. The most polluted areas, with Fe, Cr, Ni and V, of this biomonitoring resulted to be Milot, Golem, Kruje, Lezhe and Manez. The main anthropogenic sources of these elements are the windblown soil dust resulting from massive constructions, the depositions of various minerals and coal and from the agricultural activities. Other anthropogenic activities are the burning of various combustible materials and road traffic.

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