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Assessment of atmospheric pollution with heavy metals and nitrogen using *Pleurozium schreberi* mosses as bioindicator in Latvia: spatial and temporal aspects

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Abstract

Concentrations of eight metals (Cd, Cr, Cu, Fe, Ni, Pb, Zn and V) and nitrogen were determined during a nation-wide biomonitoring survey using *Pleurozium schreberi* moss in Latvia. Particular concentrations of heavy metals were clearly associated with local emission point sources in Liepāja (Cd, Cr, Cu, Fe, Pb, V, Zn), Brocēni (Ni), Riga (Cr, Cu) and Daugavpils (Cr, Cu, Fe, Ni, Zn). Increased concentrations in the western part of Latvia (Cd, Cr, Cu, Ni, Pb, V, Zn) were due to the long-range transboundary transport of pollution from Europe and local metallurgical and other factories sources in Liepāja City. Higher concentrations near the Lithuanian border were associated with the impact of pollution from the cement industry of Naujoji Akmene (Cu, Fe, Ni) and the oil refinery of Mažeikiai (Ni, V). In general, concentrations of heavy metals were lower in Latvia compared to the background levels in Europe. In comparison to the previous monitoring results, the concentration of heavy metals in moss has decreased in Latvia. The higher N concentrations in the south-western part of Latvia were due to the long-range transboundary transport of pollution, territories with intensive agriculture and areas close to the industrial cities.

Key words: air pollution, biomonitoring, heavy metals, Latvia, moss, nitrogen.

Introduction

Biomonitoring is a technique for using organisms to determine air quality change and pollution distribution. Moss is an inexpensive and effective material that accumulates atmospheric and environmental pollution deposits and, therefore, is particularly suitable for the analysis and identification of areas with high atmospheric deposition fluxes and temporal trends (Harmens et al. 2010). The recommended moss species for biomonitoring in Europe are *Hylocomium splendens, Pleurozium schreberi* (Ross 1990; Poikolainen et al. 2004; Kosior et al. 2010), *Hypnum cupressiforme* (González-Miqueo et al. 2010) and *Pseudoscleropodium purum* (Boquete et al. 2014). Mosses have been shown to be suitable as indicators of atmospheric deposition with heavy metals (Rühling, Tyler 1973; Rinne, Mäkinen 1988; Brūmelis, Nikodemus 1995; Nikodemus, Brūmelis 1998; Kayee et al. 2015), nitrogen (González-Miqueo et al. 2010; Harmens et al. 2011; Harmens et al. 2014) and persistent organic pollutants (Gałuszka 2007; Migaszewski et al. 2009; Doļegowska, Migaszewski 2011).

The moss biomonitoring method has been applied at international (Harmens et al. 2008; Harmens et al. 2013; Harmens et al. 2015), regional (Rühling, Tyler 1973; Markert et al. 1996; Čeburnis et al. 1999; Gerdol et al. 2000; Nikodemus et al. 2004; Salemaa et al. 2004; Sucharová, Suchara 2004; Zechmeister et al. 2008; Šakalys et al. 2009; Kösta, Liiv 2011) and local scales (Brūmelis, Nikodemus 1993; Nikodemus, Brūmelis 1993; Brūmelis et al. 1999; Genoni et al. 2000).

The aim of the present study was to perform a nation-wide survey on the current situation in Latvia in respect to pollution with heavy metals and nitrogen using feather moss *Pleurozium schreberi* and to compare the obtained data with the previous biomonitoring results.

Materials and methods

Moss sampling

Biomonitoring of atmospheric heavy metal distributions has been conducted using moss in five national surveys: in 1990, using *Hylocomium splendens* collected in 81 plots; and in 1995, 2000, 2005 and 2015, using *Pleurozium schreberi* in 101 plots (Fig. 1). Change from one species to the other was made because *P. schreberi* has wider distribution in Latvia. Compared with 1990, when moss was collected from pine, spruce or mixed forests, sampling in the subsequent years was made in dominant pine stands. The sampling sites in 2015 had close locations to those in 1990. Each sampling plot was located at least 300 m from major roads and at least 1 km from pollution point sources and from large cities; the plot size was 50 × 50 m. Carpet-forming bryophytes, bilberry (*Vaccinium myrtillus*) and lingonberry (*Vaccinium vitis-idaea*) dominated in the understorey. Moss sampling period was from middle August to middle October 2015.

Analytical methods

The analysis was carried out at the University of Latvia. Moss samples were dried at room temperature for about a week and then at 40 °C for 24 h, and cleaned from coniferous needles, leaves and other debris. Only green parts of the moss material were used for the analyses. All methods, including moss sampling and heavy metals analysis, were performed according to standardized methods (ICP Vegetation 2010).

Concentrations of eight metals (Cd, Cr, Cu, Fe, Ni, Pb, Zn and V) were measured using an atomic absorption spectrophotometer (AAnalyst 800, Perkin–Elmer). The modified Kjeldal method was used for the determination of nitrogen.

Quality control of the analytical process was conducted (Steinnes et al. 1997; Harmens et al. 2008). The moss reference materials M2, containing elevated concentrations of most metals, and M3, containing background concentrations of most metals, were obtained from the Coordination Centre of the International Cooperative Programme on Effects of Air Pollution on Natural Vegetation and Crops, Centre for Ecology & Hydrology, Bangor, UK.

Data analysis and mapping

Microsoft Excel was used for data entry and statistical analysis. Mapping was conducted using ArcView.

Results

Cadmium

Cadmium concentration in *P. schreberi* moss in Latvia was low (Table 1). The highest Cd concentration (0.55 mg kg^{-1}) was found in the western part of Latvia (Fig. 2A), around Liepāja.

Chromium

Due to the relatively slow development of industry, high Cr concentration (Table 1) was not characteristic in Latvia. Slightly increased Cr concentration (0.86 mg kg^{-1}) in moss was found in the western part of Latvia, around Liepāja, and this might be related to the previously mentioned factors. Slightly increased Cr concentration was also found around the largest cities of Latvia: Riga, Jelgava and Daugavpils (Fig. 2B).

Copper

Similarly to other elements, the highest Cu concentration in *P. schreberi* moss in 2015 was also found in the western part of Latvia (Fig. 2C). While the median Cu concentration in moss in Latvia was 5.17 mg kg^{-1} (Table 1), the Cu concentration in the moss collected in the western part of Latvia around Skrunda and Liepāja reached 8.95 to 12.21 mg kg^{-1}. Another separate area of Cu pollution distribution had formed in the eastern part of Latvia, around Daugavpils and Rēzekne, where the concentration of Cu reached 7.01

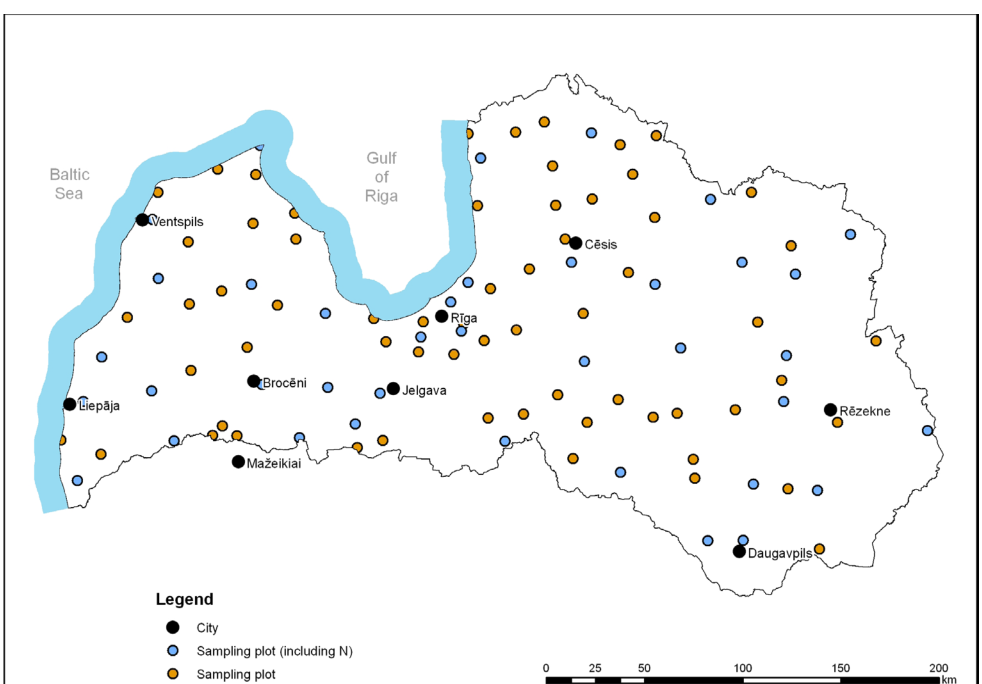

Fig. 1. Location of sampling plots in Latvia in 2015; the major cities are also shown.

Table 1. Minimum, maximum and median concentrations of heavy metals (mg kg^{-1}) and nitrogen (%) in *Pleurozium schreberi* moss in Latvia in 2015 and the median concentration in Europe in 2010 (Harmens et al. 2013)

	V	Cr	Ni	Cd	Pb	Zn	Cu	Fe	N
Minimum	0.17	0.01	0.20	0.04	0.41	22.40	2.37	36.91	0.75
Maximum	2.54	1.04	1.67	0.55	9.20	99.79	12.31	587.82	1.83
Median	0.49	0.33	0.48	0.10	1.26	33.13	5.17	133.02	1.13
Europe	1.72	1.82	1.94	0.20	3.57	31.00	6.53	538.00	1.19

and 8.18 mg kg^{-1} respectively; as well as around Riga (in Beberbeķi), with the concentration of 12.31 mg kg^{-1}.

Iron

The highest Fe concentration (329.37 to 587.82 mg kg^{-1}) in moss was found near the largest cities of western Latvia (Liepāja, Ventspils) (Fig. 2D). At the same time, attention must be drawn to the slightly increased Fe concentration around the cement industry in Brocēni (Latvia; 323.49 mg kg^{-1}) and Naujoji Akmene (Lithuania; 183.79 mg kg^{-1}).

Nickel

In 2015, an increased Ni concentration in *P. schreberi* moss in Latvia was found in small local areas (Fig. 2E). Relatively higher Ni concentration (0.97 to 1.64 mg kg^{-1}) was found in the southern part of Latvia, near the Lithuanian border. Here, the Ni concentration in *P. schreberi* moss was due to the long-range transport of pollution from the Mažeikiai Nafta oil refinery industry and the Naujoji Akmene cement industry. The increased Ni concentration around Brocēni was related to pollution emissions from the Brocēni cement industry.

Higher Ni concentration in moss was also found near other largest cities of Latvia, for example, Liepāja (Grobiņa plot, 1.00 mg kg^{-1}) and Daugavpils (Daugavpils, 0.86 mg kg-1; and Ilūkste, 1.06 mg kg^{-1}).

Lead

The highest lead concentration in *P. schreberi* moss in 2015 was found in the western part of Latvia, near Liepāja (9.20 mg kg^{-1}; Fig. 2F). Higher Pb concentration in moss was found near Kārsava (3.62 mg kg^{-1}), in the eastern part of Latvia, not far from the border with Russia.

Vanadium

Pollution with vanadium had a local character (Fig. 2G) in Latvia with an explicit decreasing tendency in the west-east direction. The highest V concentration in *P. schreberi* moss in 2015 was detected in the southern part of Latvia, near the Lithuanian border (1.42 to 2.54 mg kg^{-1}). Higher V concentration in moss was found near other largest cities: Liepāja (Grobiņa, 1.36 mg kg^{-1}; and Bārta, 1.29 mg kg^{-1}) and Ventspils (1.24 mg kg^{-1}).

Zinc

A relatively high zinc concentration in *P. schreberi* moss was found in a small area in Latvia in 2015 (Fig. 2H). For a long time, the main source of Zn pollution in Latvia has been the Liepāja metallurgical industry (48.37 to 99.79 mg kg^{-1}).

Nitrogen

The highest N concentration in *P. schreberi* moss in 2015 was identified in the south-western part of Latvia (1.74 to 1.83%), and the lowest (< 1.10%) in the northern part (Fig. 3).

Retrospective analysis of heavy metal concentrations in moss

The mapping of distribution of heavy metal pollution in Latvia using moss was performed in 1990, 1995, 2000, 2005 and 2015. Therefore, it is possible to compare these data retrospectively.

It must be mentioned that the economic situation changed significantly over this period in Latvia. When the national monitoring was started in 1990, many post-soviet industries and thermal power plants were still operating, and traffic was not intensive and mostly consisted of cars manufactured in the Soviet Union. If initially industries operated at full capacity, by 1995 many industries had already been shut down or their manufacturing intensity has greatly decreased. This is also confirmed by the fact that the concentrations of all heavy metals had decreased by 2015 in comparison to the results of 1995 (Fig. 4).

Comparison of the concentrations of all heavy metals with the average European indicators showed that the pollution level in Latvia corresponds with that in Europe, as seen by the results summarised in Table 1. Table shows that the medians of all elements, except Zn, are lower in Latvia than the average calculated median in Europe. The concentration medians of all metals are 50 % or lower than in Europe, except Zn, which is slightly higher (6.4 %).

Vanadium, chromium and nickel concentrations decreased consistently in every moss sample analysis period. Cadmium concentration levels also decreased since 1990, with an exception in 2005, when it was increased. Fig. 4 shows that pollution has decreased since 1990. However, during the last three mapping surveys of heavy metal distribution (2000, 2005 and 2015), the concentration of Cu has become stable and no concentration decrease tendency has been observed. A similar situation can be observed with Pb, as its concentration in moss has not decreased significantly since 2000. Further, in comparison with 1990,

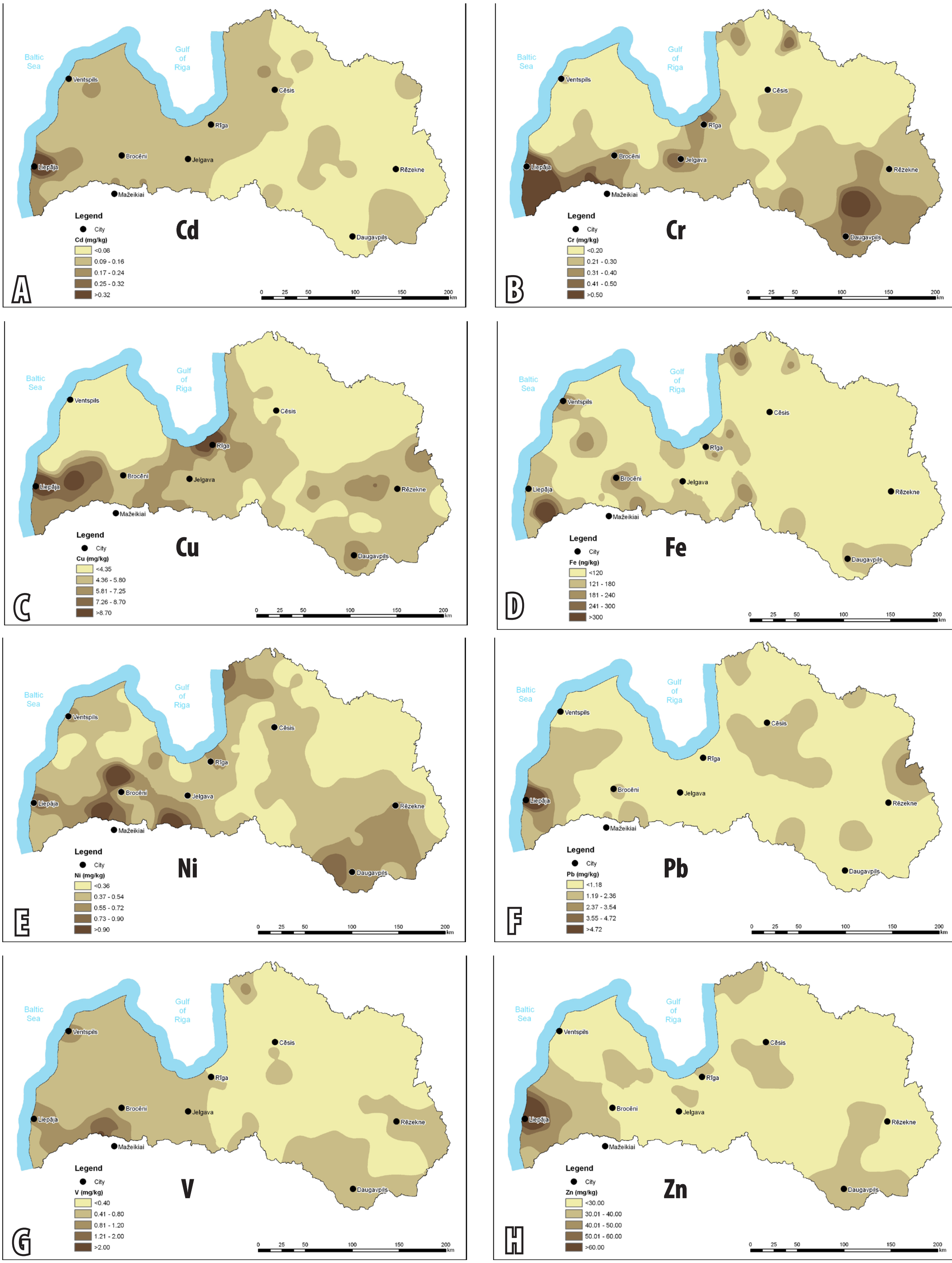

Fig. 2. Heavy metal Cd (A), Cr (B), Cu (C), Fe (D), Ni (E), Pb (F), V (G), Zn(H) concentrations (mg kg^{-1}) in moss *Pleurozium schreberi* in Latvia in 2015.

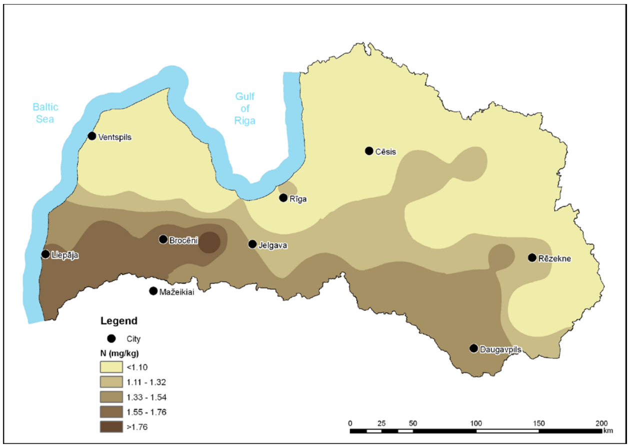

Fig. 3. Nitrogen concentration (%) in moss *Pleurozium schreberi* in Latvia in 2015.

the data of 2015 show a decrease in Zn concentration. However, the data from 1995 through 2000 (Fig. 4) reveal an unchanged concentration over this period. Next, the overall Fe concentrations in Latvia have decreased since 1995, while there have been no radical concentration shift since 2000 (Fig. 4).

Discussion

In Latvia the overall concentrations of heavy metals in mosses at all plots in the period from 1990 to 2015 have declined the most for lead (89%), vanadium (85%), chromium (78%), iron (71%), cadmium (69%) and nickel (66%), followed by zinc (21%) and copper (14%). Maps of heavy metal distribution (Fig. 2), clearly indicate that local pollution zones in Latvia are divided into the Liepāja (Zn, V, Pb, Fe, Cu, Cd, Cr), Ventspils (Fe, V), Brocēni (Fe, Zn, Ni, Cu), Riga (Cu, Cr), Daugavpils (Cu, Ni, Fe, Zn, Cr), Mažeikiai (V, Ni) and Naujoji Akmene (Cu, Fe, Ni) areas. There are three main factors that stand out, explaining some of the pollutants and their distribution tendencies: windblown dust, long-range transport of air pollution and local pollution sources (Harmens et al. 2015). The presence of some other factors also cannot be ruled out.

Notably, the most complex heavy metal pollution

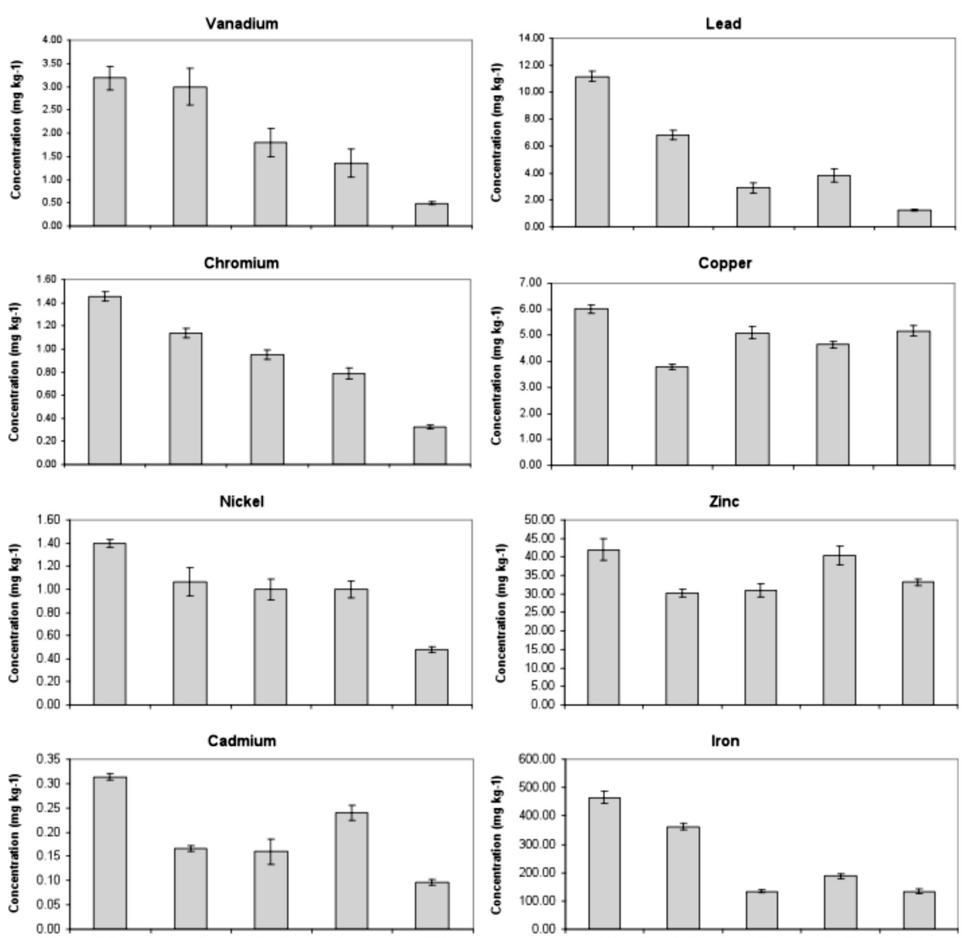

Fig. 4. Median heavy metal (V, Cr, Ni, Cd, Pb, Cu, Zn, Fe) concentrations (mg kg^{-1}) in mosses in Latvia in five surveys from 1990 to 2015 (1990, 1995, 2000, 2005, 2015). Standard errors are shown.

is characteristic to the Liepāja area, mainly caused by the operations of metallurgy industries in Liepāja and long-range transport of pollution from Western Europe (Nikodemus, Brūmelis 1998, Nikodemus et al. 2004).

Decrease in V and Ni concentrations (Fig. 4) can be explained by the fact that many thermal power plant have substituted oil products with gas or biofuel (Nikodemus et al. 2004) that are more environmental friendly. Similar conclusions were made in Lithuania: when oil was used as fuel in the Naujoji Akmene plant, a higher pollution level of Ni was identified around it, and increased pollution with V was found close to the Alytus thermal power plant (Čeburnis et al. 1997). Kvietkus et al. (2011) also note that anthropogenic elements (Pb, Zn, Cr, Ni, Cu, Cd) have higher concentrations during the cold period, which is indicative of more intense operations of thermal power industry during the heating season. Compared to 2000 (Nikodemus et al. 2004) and 2005 (Harmens et al. 2008), the maximum V concentrations in moss near Ezere plot (Lithuania border) have decreased more than nine times. Previous mappings do not show increased V concentrations in moss near Riga (Nikodemus, Brūmelis 1998, Nikodemus et al. 2004), and this can be related to the switching from liquid fuel to gas in the Riga Thermal Power Plant 2 (TEC 2). Similarly to V, Ni can be related to the operations of the Mažeikiai Nafta oil refinery plant.

Lead concentration in moss in the vicinity of Liepāja in 2015 decreased 5.4 times when compared to 2005. This decrease might be related to the irregular operations of the most important environment polluter of that area – the metallurgy industry of Liepājas Metalurgs AS, as well as to the decrease in long-range transport of pollution from Western Europe. Pb concentration in moss depends on traffic intensity (Kösta, Liiv 2011). In addition, decrease of Pb concentrations could be related not only to the decline in the intensity of industrial manufacturing but also to the improved emission control not only in Latvia but also in entire Europe, as well as to the decrease of usage of Pb-containing fuel and improvement of road transport quality (EEA 2012).

Copper is one of the heavy metals whose concentration in *P. schreberi* moss has not decreased during the last decade in Latvia. Oil products contain high concentrations of Cu, and road transport, metallurgical industry and burning of fossil fuels also account for pollution with this heavy metal (Harmens et al. 2013). Moreover, for many years, pharmaceutical companies have been significant sources of Cu pollution in Latvia, especially in Olaine (Brūmelis, Nikodemus, 1993). Now, as shown in Figure 2C, the most significant sources of pollution are located in industrial cities of Latvia (Liepāja, Riga, Rēzekne, Daugavpils) as well as in Brocēni and Naujoji Akmene (cement industries). The locations of pollution sources have not changed when compared to the previous stages of heavy metal distribution mapping (Nikodemus et al. 2004).

Iron concentration in *P. schreberi* moss and its territorial distribution in Latvia is related to both natural and anthropogenic factors. Concentrations of elements of natural origin (Fe and Mn) were higher during the warm period (Kvietkus et al. 2011), and this is related to iron pollution entering the environment due to industry. In agricultural areas, in turn, the source of Fe pollution can be soil dust (Harmens et al. 2007). Fe concentration decrease in moss in Latvia can be explained by the change from oil and coal fuel to biofuel in thermal power plants and cogeneration stations (Central Statistical Bureau 2016) and from liquid fuel to gas in Riga TEC 2, as well as by the decrease of arable land areas (Silava 2016).

Increased Cr concentrations in moss in Latvia are focused around industrial cities: Liepāja, Olaine, Riga and Daugavpils. Generally, higher Cr levels are found in the southern part of Latvia (Fig. 2B). Starting with 1990, a steady decreasing tendency in Cr concentration in moss can be observed in the entire territory of Latvia and especially around Brocēni. Also around the Kunda Nordic Tsement AS cement industry in NE Estonia, the concentration of Cr has decreased by 59% over the period of 1989–2008 (Kösta, Liiv 2011), while its concentration around the Latvian cement industry in Brocēni has decreased by 71% over the period from 1990 to 2005 and by 84% from 1990 to 2015. The decrease of Cr concentration can be explained by a rapid reduction in the operational intensity of heavy metallurgy and cement manufacture industries. Like Cr, Cd is also widely used in heavy metallurgy industries (EEA 2012). Cd pollution has significantly decreased in Latvia and in entire Europe (Harmens et al. 2010).

Most metallurgical industries emit Zn in the atmosphere (Harmens et al. 2007); this is also the case in Latvia, around the Liepāja metallurgical industry in particular. In comparison with 2000, when the maximum Zn concentration in Latvia in the direct impact zone of the Liepāja metallurgical industry was 189.00 mg kg^{-1} (Nikodemus et al. 2004), by now the concentration of Zn in moss has dropped to 99.79 mg kg^{-1} – almost twice over the last 15 years. This drop is definitely related to the economic difficulties and irregular operations of Liepājas metalurgs AS.

On the European scale, the median N concentrations in 2005 and 2010 were 1.26 % and 1.19 % respectively (Harmens et al. 2013). According to our results, the median N concentration in 2015 was 1.13 % (Table 1). This roughly corresponds to the background level in European countries.

The highest N concentrations are found in the south-western part of Latvia, due to the long-range transboundary transport of pollution, as well as in the territories with intensive agriculture and areas close to industrial cities. Generally, mosses affected by canopy drip have higher N concentrations due to the higher N content in throughfall deposition (Skudnik et al. 2014). Nitrogen presented a more diffuse spatial distribution in the entire area.

Generally, when assessing the situation in Latvia, it can be noticed that the atmospheric pollution with heavy metals is not high and has a decreasing tendency at regional levels. At the same time, in the future, attention should be paid to local pollution sources in cities where bioindicators can be used for determining atmospheric pollution (Pīrāga et al. 2017).

The mapping results of heavy metal distribution in the atmosphere in Latvia in 2015 using *P. schreberi* moss confirmed its efficiency in atmospheric air quality change control. This method allows to determine not only spatial heavy metal and nitrogen distribution but also temporal changes.

Acknowledgements

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