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Bio-monitoring of Heavy Metals in the Vicinity of Copper Mining Site at Erdenet, Mongolia

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ABSTRACT
The objective of this research was to evaluate the level of heavy metal concentrations of copper, lead, zinc, cobalt, manganese, iron and nickle (Cu, Pb, Zn, Co, Mn, Fe and Ni) in correlation with human activities and land use at different sites of Erdenet, Mongolia. Samples of tree bark, lichens and street dust were collected and analyzed form the main highway, road intersections, residential areas, remote sites and the vicinity of the copper mine area near the city of Erdenet, Mongolia. The copper levels in all sampling sites with respect to all media differ from each other consistently and are clearly related to the distance from the copper mine. This indicates that the mining activities are the main source of copper rather than other anthropogenic activities in Erdenet, Mongolia. The lowest metal concentrations were found in residential areas. For street dust samples higher metal concentrations were found in the fine grains particles, which were attributed to higher surface area. This research provided strong evidence of air pollution as it could be concluded that tree bark and lichens can be recommended as efficient bioindicators of heavy metals air pollution in cold climate regions.

Key words: Mongolia, copper mining, bioindicator, heavy metals, street dust, lichens, tree bark

INTRODUCTION

Metals are non-biodegradable and persist for long periods in terrestrial environments. They may be transported through air and water to reach surface and ground waters or may be taken up by plants, including agricultural crops. Accumulation of heavy metals in crops grown in metal-polluted soil may easily cause damaging effects on human health through ingestion (Ziadat *et al*., 2006; Balabanova *et al*., 2010; Garty and Garty-Spitz, 2015).

Fu *et al*. (2008) conducted an investigation of heavy metal content in rice sampled from Taizhou in Zhejiang where elevated levels of heavy metals were reported.

Mining activities are one of the major sources of heavy metal atmospheric releases around the world. There are different sources of metal contamination in mining areas, including grinding, concentrating ores and tailings disposal (Garty and Garty-Spitz, 2015; Rastmanesh *et al*., 2011; Wang *et al*., 2004). Inappropriate treatment and disposal of tailings and acid mine drainage could pollute agricultural fields surrounding the mining areas (Williams *et al*., 2009).

Mining generally releases toxic heavy metals such as copper (Cu), arsenic (As), lead (Pb) and mercury (Hg) by excavation of the mine area for mining process. The adverse effects of mining activity on the environment as well as human health have been observed in many areas (Mandiwana *et al*., 2006; Olias *et al*., 2004; Murao *et al*., 2002; Martin *et al*., 1998).

Copper is one of the numerous metals significantly toxic to human beings and ecological environments (Gorell *et al*., 1999). Metal mining and smelting have been recognized as major contributors to environmental pollution over the past few decades, frequently acting as sources of heavy metals (Ettler *et al*., 2011; Benvenuti *et al*., 1997; Gray, 1997; Liu *et al*., 1999). In the past decade, numerous studies of heavy metal contamination in areas surrounding mining operations have been completed. Jiries *et al*. (2003) reported elevated Pb concentration in the effluent water produced from phosphate mining in Jordan. El-Hasan (2006) reported that a large portion of the heavy metals released are concentrated in the slime associated with phosphate wash at Al-Abyad mine in Jordan. High concentrations of Hg and Cu were found in the Boroo River, which flows through gold deposit areas in the Northern part of Mongolia (Tumenbayar *et al*., 2000; Inam *et al*., 2011).

There are multiple ways to investigate heavy metals atmospheric pollution with bio-indicators including the following: suspended dust (Jiries *et al*., 2008), dry deposition (Sawidis *et al*., 2012; Ziadat *et al*., 2006), wet precipitation (Jaradat *et al*., 1999), plants (Turan *et al*., 2011; Jiries *et al*., 2008) and lichens (Lisowska, 2011; Berdanier *et al*., 2009). Heavy metal concentrations are known to reach high levels in lichens when grown on metal-rich substrates. Very little attention has been given to the impact of mining activities on air pollution in Mongolia. A study by Hauck (2008) indicated a general increase in air pollution in the Mongolian capital Ulan Bator by using epiphytic lichens as bioindicators.

The aim of this work is to investigate the air quality in terms of heavy metal concentrations in areas surrounding the Erdenet copper mine using street dust, tree bark and lichens as bioindicators.

MATERIALS AND METHODS

Research area: Mongolia is situated in the central part of the Asiatic continent. The country is bounded on the North by Russia and on the East, South and West by China (Fig. 1).

Erdenet is a small industrial city located in Western Mongolia at an elevation of 1580 m amsl, characterized by high annual sunshine exposure, short dry summer months and long cold winter season climatic conditions. Erdenet is the second largest industrial and mining city in Mongolia with a population of 100 thousand and one of the largest open pit copper mines in the world.

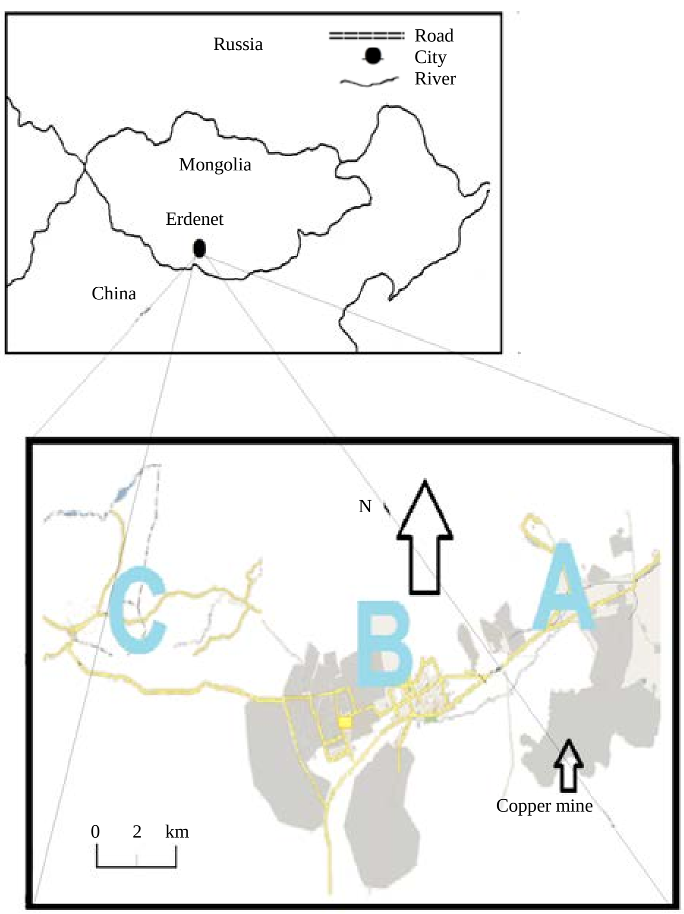

Fig. 1: Map of sampling locations in the vicinity of the city of Erdenet, Mongolia

Erdenet copper mine has been in operation since 1978, at a production capacity of approximately 20 million Mt of ore annually, which yields approximately 354,000 Mt of copper concentrate each year (MBendi, 2015).

The main pollution sources of heavy metals and trace elements in the Erdenet mining environment is the white dust from the storage of slag-heap tail ores of the open-pit mine and the storage of nonstandard ores, rocks and rubbish. Around 25 million tons of tail pulps are currently disposed every year to the storage in slag-heap tailing ponds (Gerbish *et al*., 1993).

Sample collection: In order to identify the copper mine activities as a potential source of different heavy metal pollution in the area, selected heavy metal concentrations were determined from street dust, tree bark and lichens at various sites within the city of Erdenet. Sampling areas were divided into three different categories based on the distance from the copper mine and the traffic density in the area as shown in Fig. 1:

- **Area (A):** Samples collected from locations surrounding the copper mine activities
- **Area (B):** Samples collected from high traffic density areas
- **Area (C):** Samples collected from low traffic density areas

Samples from a remote area (50 km from Erdenet) were also collected and analyzed for background reference concentrations for comparison.

In-situ samples of street dust, larch tree bark (*L. occidentalis*) which is widely spread in the investigation area and healthy lichens from rock surfaces at a height >1.5 m above the ground were collected from 18 locations in Erdenet. Road sediment samples from the road sides were collected with a plastic scoop from three areas within Erdenet representing the above mentioned sites during the month of July 2008.

Nine sampling sites from the city center and nine sampling sites from residential areas were chosen. In addition, three samples from each sampling media collected from a remote site, characterized with similar climate, altitude and lithology to be used as background samples.

Three replicates from each type of media sampled were collected from each site to get a representative sample. The selected heavy metals used in this investigation (Cu, Pb, Zn, Co, Mn, Fe and Ni) were based on their toxicity and possible sources of metal pollution.

Sample analysis: Directly after collecting hydrated lichens and tree bark, the samples were transferred to the laboratory and washed with distilled water for approximately 1 min to remove any collected dust on the lichen surface that might interfere with the analysis. Elemental composition of unwashed samples are affected by street dust contamination, thereby leading to incorrect interpretations of baseline concentrations and relationships between elements (Loppi *et al*., 1999). Washing the samples should not have any effect on the trace metal content as the metals are more tightly bound or sequestered within lichens and therefore more slowly released (Garty, 2001). Washed samples were dried at room temperature overnight and then oven-dried to a constant weight at 80°C. The dried lichen samples were then prepared for metal analysis after being ground, using an agate mortar to avoid metal contamination.

Sediment samples were divided into erinaceous (63-2000 mm) and argillaceous (<63 mm) fractions. A previous method used by Jiries (2003) was applied for street dust digestion. Samples (0.5 g) of dried lichen, street dust and tree bark were digested in a mixture of concentrated HNO_3 of 70% purity and $HClO_4$ of 98% purity (2:1, v/v) at 80°C using heating blocks overnight. The samples were filtered through Whatman filter paper No. 42 from England and diluted to 25 mL with deionized water. The total concentrations of Cu, Pb, Zn, Co, Mn, Fe and Ni in the samples were determined using a PerkinElmer atomic adsorption spectrophotometer model Analyst 300 equipped with a graphite furnace model HAG 800 and an AS-72 autosampler (PerkinElmer, Germany). The concentrations of trace metals were measured using external calibration of multi-elements standard solution, where r^2>0.9995.

The analysis was carried out in triplicate and the average values were reported. Instrument precisions were determined by introducing the same quantity of one sample 20 times and then the relative standard deviation was calculated (RSD <5%).

RESULTS AND DISCUSSION

Heavy metals in street dust: The samples were analyzed according to grain size distribution (fine <63 mm and coarse 63-2000 mm). Concentration of heavy metals (Cu, Mn, Zn, Ni, Co, Fe and Pb) for both fine and coarse grain sizes were collected from street dust of different locations within Erdenet are presented in Table 1.

It is worth mentioning that copper concentration was the dominant heavy metal of both fine and coarse grain size among all analyzed heavy metals after iron which highlights the direct impact of copper mining activities on the investigated sites surrounding the copper mine area, this is in full agreement with previous researches conducted by Balabanova *et al*. (2010) and Garty and Garty-Spitz (2015).

The higher concentration of Cu in fine particles at sites B and C can be attributed to the fact that fine particles are more

Table 1: Statistical summary of heavy metal concentrations in for street dust samples collected from different areas within Erdenet

Grain size and heavy metal	Area A				Area B				Area c			
	Min.	Max.	Mean	STD	Min.	Max.	Mean	STD	Min.	Max.	Mean	STD
Coarse size (mg kg^{-1})												
Pb	7.0	10.5	9.3	±0.5	8.4	16.9	12.6	±3.3	7.2	16.9	10.8	±3.7
Co	6.2	10.6	8.1	±1.2	5.6	9.4	5.9	±1.4	3.0	9.4	5.6	±1.0
Ni	2.8	4.4	3.3	±1.4	5.8	12.1	7.2	±2.2	3.6	12.1	5.8	±1.7
Zn	2.6	175.0	126.2	±60.4	79.6	101.6	77.7	±27.2	64.3	111.2	79.6	±29.5
Fe	25755.0	40175	33087.5	±8090.3	16115.1	22200.5	16655.1	±3101.1	11421.8	22200.5	16115.1	±2257.6
Mn	172.1	260.0	217.2	±57.7	234.6	326.3	266.6	±41.6	160.0	326.3	234.6	±43.0
Cu	558.0	1535.0	1145.1	±692.6	244.0	326.0	179.7	±90.9	176.3	326.0	244.0	±67.1
Fine size												
Pb	31.7	47.0	39.8	±8.2	23.6	40.4	29.9	±9.6	8.7	17.3	15.7	±5.9
Co	15.7	19.4	18.0	±3.4	11.3	16.2	13.2	±3.3	7.9	13.9	10.6	±1.4
Ni	10.6	12.1	13.7	±2.4	14.4	20.1	16.5	±3.9	10.2	19.7	12.8	±2.0
Zn	269.7	296.0	264.1	±76.9	122.6	198.1	149.5	±45.9	76.2	119.9	97.7	±15.5
Fe	55515.6	62942.7	59925.8	±11051.9	3250.0	47687.5	31.862.5	±17448.3	25281.2	42812.2	31703.1	±2793.4
Mn	395.6	415.4	405.8	±63.2	409.5	525.1	479.8	±85.0	331.4	530.0	428.5	±44.9
Cu	2810.2	4740.9	4026.8	±2261.6	233.2	1927.6	727.3	±669.5	266.3	499.2	365.8	±88.7

Min: Minimum, Max: Maximum, STD: Standard deviation

Table 2: Correlation coefficient for different heavy metals in 18 street dust samples collected from different areas within Erdenet

Heavy metals	Pb	Co	Ni	Zn	Fe	Mn
Co	0.75					
Ni	0.53	0.69				
Zn	0.79	0.82	0.66			
Fe	0.68	0.78	0.46	0.79		
Mn	0.53	0.76	0.86	0.45	0.48	
Cu	0.72	0.71	0.17	0.91	0.80	0.20

easily transported by wind than coarse particles in addition to higher surface area of the fine particles. However, the copper level nearest to the mining site was above the presumable natural level of street dust reported by Cassella *et al*. (2007) reaching 40 mg kg^{-1} as this can be attributed to the topography of the surrounding areas of the copper mine.

The concentrations of Pb, Mn and Ni in both fine and coarse size particles varied significantly according to traffic density, as the concentrations in area B > area C > area A. This result is due to the fact that the main sources of these elements are from traffic emission rather than from mining activities in the investigated area.

The concentrations of Co, Zn and Fe were highest in area A and the lowest in area C. Such elements could be directly associated with emissions from the mining activities.

The results of the sampling analyses from street dust showed higher concentration of Cu, Fe and Zn in area A, in comparison to the area B and area C, respectively as shown in Fig. 2 and were higher than the background samples. The results were attributed to mining activities of the ore during ore excavation and handling processes.

The copper levels in the above mentioned areas differ from each other consistently and are clearly related to distance from the copper mine. This indicates that the mining activities are the main source of copper rather than other anthropogenic activities which in full agreement with Ettler *et al*. (2011).

The heavy metal concentrations of all analyzed samples showed higher concentrations in fine portions of the samples rather than the coarse portions as shown in Fig. 3 which is attributed to higher surface area of the fine particles. On the other hand, the lead level recorded in the high traffic density areas, 8.4-16.9 μg g^{-1} for coarse particles and 23.6-40.4 μg g^{-1} for fine grained particles, is low compared to values reported for street dust along busy roads in other countries such as Amman-Jordan (Jiries, 2003). This can be attributed to lower number of vehicles in Erdenet compared with other countries.

Correlation analysis was carried out to determine the extent of the relationship between the investigated heavy metals (Table 2). The correlation matrix shows significant correlation for Cu with Pb, Co, Zn and Fe (r between 0.71 and 0.91), for Ni with Mn (r = 0.86) and for Co with Ni, Zn, Fe and Mn (r between 0.69 and 0.82) in samples taken from street dust of Erdenet suggesting a common source for these metals in the environment most probably from the mine dust deposited on the investigation site in addition to vehicle emission. The moderate correlations among other metals indicated multi and combined-sources of pollution.

Heavy metals in tree bark: The concentrations of heavy metals in tree bark originates mainly from atmospheric particulates rather than the street dust as the particulates are included within the trunks of tree bark pockets (Sawidis *et al*., 2012; Bellis *et al*., 2003; El-Hasan *et al*., 2002; Al Alawi *et al*., 2007).

The concentration of heavy metals in tree bark samples from different locations of Erdenet are presented in Table 3. Copper showed the highest concentration after iron among analyzed heavy metals which can be attributed to copper released into the atmosphere during mining excavation.

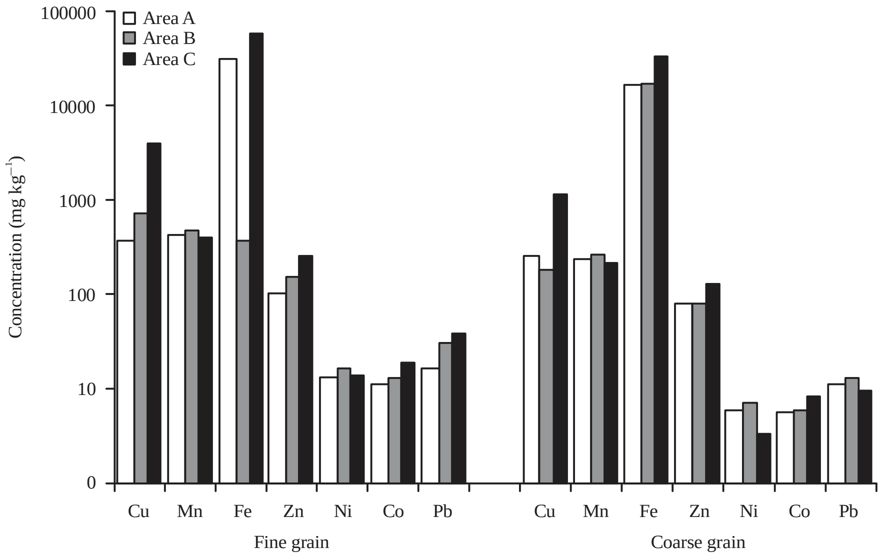

Fig. 2: Heavy metal concentrations of fine and coarse grained particles in street dust samples collected from different areas within Erdenet

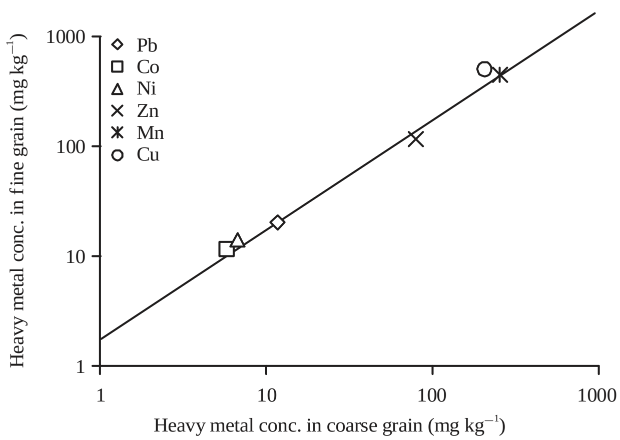

Fig. 3: Heavy metal concentrations for fine particles compared with coarse grain particles of the same samples

Table 3: Statistical summary of heavy metal concentrations in tree barks from different areas within Erdenet

	Heavy metal concentration (mg kg^{-1})						
Area and parameters	Pb	Co	Ni	Zn	Fe	Mn	Cu
Area A							
Min.	0.5	6.9	2.0	8.4	563.0	17.8	84.5
Max.	2.3	9.7	3.8	82.5	1061.3	26.1	146.0
Mean	1.1	7.1	2.7	28.9	739.2	21.2	125.5
STD	±0.6	±1.4	±0.8	±7.8	±222.1	±3.6	±35.7
Area B							
Min.	0.6	8.8	0.1	3.8	22.3	22.3	107.3
Max.	3.7	6.1	5.7	53.5	1577.5	80.1	123.8
Mean	1.4	7.1	2.3	16.2	631.7	48.8	113.3
STD	±0.3	±0.4	±1.8	±31.7	±456.7	±15.6	±6.1
Area C							
Min.	0.4	2.4	0.1	3.3	31.7	10.4	54.7
Max.	0.7	3.3	4.9	25.0	1361.0	61.5	162.5
Mean	0.5	2.8	2.2	11.6	614.1	31.5	106.0
STD	±1.2	±0.9	±1.8	±22.5	±574.0	±22.7	±17.3

Min: Minimum, Max: Maximum, STD: Standard deviation

Table 4: Statistical summary of heavy metal concentrations in lichens from high and low traffic densities within Erdenet, no lichens were found in area A

	Heavy metal concentration (mg kg^{-1})						
Area and parameters	Pb	Co	Ni	Zn	Fe	Mn	Cu
Area B (N = 6)							
Min.	0.3	0.5	0.4	3.5	1084.4	16.9	22.5
Max.	1.0	0.6	0.5	14.4	1851.2	19.7	33.1
Mean	0.7	0.6	0.4	8.3	1286.2	18.6	27.5
STD	0.6	0.2	0.3	30.3	589.6	9.6	11.0
Area C (N = 6)							
Min.	0.4	0.5	0.2	3.5	934.4	16.3	3.1
Max.	1.9	0.6	0.7	82.6	1463.1	27.6	16.1
Mean	1.2	0.6	0.6	32.7	1143.7	22.8	8.9
STD	0.1	0.1	0.1	1.4	316.9	1.3	4.8

Min: Minimum, Max: Maximum, STD: Standard deviation

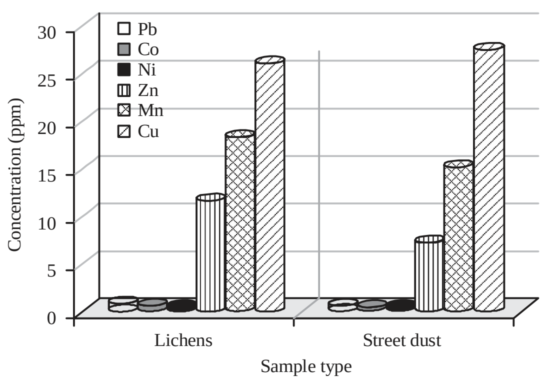

Fig. 4: Average heavy metal concentrations in lichens and street dust of the investigation site

Similar increase in copper concentrations in tree bark due to copper mining activity at Ashio copper mine and smelter in Japan was observed (Bellis *et al*., 2003). The trend of heavy metal concentrations in tree bark was similar to that of street dust (Fig. 4) indicating that the source is the same of ambient dust emission from mining activities.

The highest concentration of Cu, Fe and Zn were found in area A and the lowest in area C indicating that the area surrounding the mine is highly polluted. On the other hand Mn concentration was highest at area B which is due to traffic emission rather than mining activities as recent use of Mn-containing additives as a substitute for Pb in gasoline may eventually result in automotive Mn emissions (Ardeleanu *et al*., 1999). The major source of Ni, Co and Fe could be from fuel combustion as elevated levels of nickel concentrations were found in Netherland mosses containing up to 20 mg kg^{-1} (Kuik and Wolterbeek, 1995).

Heavy metals in lichens: The concentrations of heavy metals in collected lichens shown in Table 4 were much higher than background samples. The accumulation of Cu ranged from 22.5-33.1 mg kg^{-1} in area B and from 3.1-16.1 mg kg^{-1} in area C. These results are in agreement with copper concentrations in mosses in the Republic of Macedonia which was reported to range between 2.1-198 mg kg^{-1} with a median value of 22 mg kg^{-1} (Balabanova *et al*., 2010). However, no lichens were available at area A, near the mining area in the Erdenet. The total number of lichens species were observed to increase with increasing distances from the center of the mine.

Although, the copper mine influenced the whole area through windblown sediments and traffic density contributed additional copper to street dust through corrosion of vehicle parts. Therefore copper concentration was higher at area B than area C indicating that additional source of Cu at area B areas is most probably due to traffic rather than mining activities.

The highest concentrations among analyzed heavy metals were Cu after Fe due to atmospheric pollution caused by copper mining activities. The average concentration of heavy metals in lichens samples showed the same trend as dust samples (Fig. 4) indicating the source of heavy metals in lichens originates from ambient dust. Similar results were reported in copper mines in Greece where a significant correlation was found between the copper content in the street dust and that of the lichens *thalli* (Chettri *et al*., 1997).

CONCLUSION

The results obtained from this research indicated that the area surrounding the copper mine in Erdenet, Mongolia is polluted with heavy metals especially copper. This is attributed to the dust emission from mining activities in addition to vehicle emission in the area. Higher concentrations of heavy metals in street dust, tree bark and lichens were found near the mining area in comparison to samples from other sites. This reflects the fact that mining activities had a direct impact on atmospheric pollution with heavy metals in Erdenet, Mongolia. Chemical analysis and assessment of heavy metal concentrations in lichens and trees bark is proven through this research to be a valuable method to evaluate the atmospheric quality of heavy metals. The analysis of samples from lichens, tree bark and street dust directly confirmed the results and provided strong evidence of air pollution. Similar trends for heavy metals distribution in street dust and lichens indicated the main source of heavy metals in lichens is the ambient dust in the study area.

Taking into account the results obtained in the present research, tree bark and lichens can be recommended as efficient bio indicators of heavy metals air pollution in cold climate regions.

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