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Analysis of Moss and Topsoil to Monitor Metal Emissions from a Pulp and Paper Mill in Western Kenya

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Abstract The mean concentrations of Zn, Cu, Cd, Cr and Fe in mosses around Webuye Pan Paper factory were found to be significantly higher than in controls taken from Kakamega, indicating atmospheric contribution from the anthropogenic activities within the factory, shown within a radius of 1 km around the factory. The mean concentrations (μg/g dry weight) in the mosses ranged from 60.9–124.5, 22.6–34.2, 13.3–14.7, 20.1–33.3 and 1,822.4–2,079.2, respectively. The mean concentrations of Pb in mosses sampled from the eastern and western side of the factory were also significantly higher than in controls. The mean concentrations of Zn, Cu, Cd, Cr, Pb and Fe in topsoil samples ranged from 7 (Pb)–2,310 (Fe) mg/kg dry wt, depending on sample means based on site direction, distance from the factory and season. There was no clear variation in metal concentrations in mosses and topsoils with direction from the factory which was consistent with the observed fluctuations in wind direction which changed from south-west in the mornings to north and south-west in the afternoons during sampling.

Keywords Heavy metals · Factory emissions · Moss · Surface soil

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The use of mosses as biomonitors of airborne metal deposition has been widely reported since the 1960s and standard procedures for sampling and analysis developed in various environments worldwide (Buse et al. 2003; Krishna et al. 2003; Pepi et al. 2006; Belivermis et al. 2008; Saxena et al. 2008; UNECE 2008). Mosses, like other bryophytes, accumulate trace elements in their tissues due to lack of root, cuticle, vascular and epidermis systems, and have several advantages as biomonitors which include their higher surface area/mass ratios and higher cation-exchange capacity compared with higher plants (Markert et al. 1999; Adamo et al. 2007). Using mosses as biomonitors helps to determine the type, quantity and sources of atmospheric pollutants. Two moss species are favoured, *Pleurozium schreberi* and *Hylocomium splendens* but other species such as *Scleropodium purum* and *Thuidium tamariscinum* might be necessary, depending on the country and species locally available (UNECE 2008). Analysis of topsoil has also been found to be of significant importance in atmospheric metal pollution monitoring because it is a major recipient of atmospheric pollutants (Singh 2001; Belivermis et al. 2008). In fluvial environments, heavy metal pollution often result from direct atmospheric deposition emanating from geologic weathering or through the discharge of agricultural, municipal, residential or industrial waste products (Wagner and Boman 2003).

Heavy metal contamination of the biosphere has increased sharply since 1900 because metal residues can accumulate in the environment over long periods of time (UNECE 2008). The knowledge of the sources and fate of heavy metals in the environment is important because they pose increasing environmental and food chain problems. The Pan African Paper Mill in Webuye, where the reported study was done, is situated in an environmentally sensitive and densely populated area on the banks of River Nzoia, which flows into Lake Victoria. The factory belches out smoke, effluent and sludge thereby polluting air, soils and water in the nearby rivers, particularly River Nzoia which is the recipient of its effluent. The gaseous emissions from the factory and consequent metal depositions are considered to be the cause of rusting and other forms of corrosion which is prevalent in the area (Oduor 1994). Burning of industrial oil, biomass fuels and solid waste as well as activities in the vehicle repair workshop within the factory premises are believed to release metals into the environment. The environmental pollution caused by this mill is believed to be partly responsible for a number of health problems in the area where more than 60% of the children, between the ages of one to 5 years, born after 1974, when the plant began to operate, have had breathing problems (Oduor 1994). The specific sources and levels of contamination are not known. However, pulp and paper factories worldwide are, known to cause environmental pollution problems especially in rivers which receive their effluents (Parrott et al. 2006). The objective of this study was to determine the total concentration levels of heavy metals Pb, Cu, Cr, Zn, Fe and Cd in topsoil and moss around the Pan Paper manufacturing factory in order to evaluate the contribution of the pulp and paper factory to heavy metal emissions and loads.

Materials and Methods

The study was carried out in two locations in western part of Kenya i.e. in Kakamega forest (a control site) at 1,788 m altitude, located at latitude 0°, 41′ South and longitude 34°, 48′ East and Webuye (within 1 km radius from the Pan Paper factory) in Bungoma at 1,788 m altitude, 0° 4′ 55″ North latitude and 35° 35′ 41″ East longitude. The weather data for the two sampling areas including air temperature, wind speed, percent relative humidity and rainfall were recorded from April to June 2007 prior to the wet season sampling and are reported.

Moss (the dense carpet form of *Hylocomium splendens* species) and surface soil samples were collected from twelve sites in Webuye, i.e. three sites 1, 2 and 3, located around the Pan Paper factory at 1 km, 500 m and 100 m from the factory, respectively, in each of the four directions (i.e. south, west, east and north, respectively) relative to the factory. Sampling was conducted in two seasons—wet (June 2007) and dry (January 2007). The moss and topsoil samples were marked as N1, N2, N3, S1, S2, S3, W1, W2, W3, E1, E2 and E3 corresponding to the sampling areas falling on three circular rings around the Pan Paper factory, where N, S, W and E represent northern, southern, western and eastern directions from the factory and 1, 2, and 3 represent 1 km, 500 m and 100 m distances from the factory, respectively. For mosses, each sampling point was 3 m away from the nearest trees, without possible pronounced influence of canopy drip from trees, and preferably on the ground or on the level surface of decaying stumps, as part of the recommended precautions (UNECE 2008). A composite sample, consisting of five to ten subsamples, of moss and topsoil (0–15 cm layer), respectively, was made from each sampling point. The moss samples were picked by hand using plastic gloves at different distances, i.e. within a distance of 100 m, 500 m, and 1 km, respectively, from the factory. The composite samples were placed in plastic polythene bags, labeled and carefully closed to prevent contamination during transportation to the laboratory. Control samples (in triplicates) of moss were collected from Kakamega Forest, which has similar climate patterns as Webuye area, but is located far from the factory. In general, the direction of the wind around the factory was south-west in the morning and north and south-west in the afternoon.

Foreign materials adhering to the surface of the moss samples such as bits of tree barks, lichens, soil, dust and detritus were removed carefully in dry condition. The greenish-brown part of the moss plant representing the last 2–3 years of growth was used in this analysis (UNECE 2008). After removal of foreign materials, the moss samples were dried at 40°C for 48 h in a Gallenkamp oven until constant weights were attained. Samples of 1.0 g (dry weight) were crushed using a pestle and a mortar. The ground samples were all digested with 6 mL of concentrated nitric acid, per sample, at 120°C for 6 h. After cooling to room temperature, the samples were filtered through 0.45 μm polyethersulfon membrane filters and distilled water added up to a volume of 50 mL.

The samples were analyzed by an atomic absorption spectrophotometer, a Shimadzu AA-6300 model, for Cu, Zn, Pb, Cr, Cd and Fe at 324.8, 213.9, 217, 357.9, 228.8 and 248.3 nm, respectively, using an air-acetylene flame and quantification done using calibration standards. For data analysis, the moss data set was arranged in a matrix with rows and columns, where the rows corresponded to the sampling site and the columns to the six heavy metal concentrations recorded in the moss samples. In this case, S_1, S_2 and S_3 represented the three different radii, 100 m, 500 m and 1 km from the Pan Paper factory and triplicates were taken from every sampling point with $s_{1,1}$, $s_{1,2}$, $s_{1,3}$, $s_{2,1}$, $s_{2,2}$, $s_{2,3}$, $s_{3,1}$, $s_{3,2}$, $s_{3,3}$ representing the triplicates sampled at 100 m, 500 m and 1 km distances, respectively. Samples were analyzed in triplicates and means and standard deviations computed. The results were subjected to regression analysis and ANOVA for statistical analysis.

The soil samples, taken from the same sites as the moss samples, were air-dried for 2 days in the laboratory (21–22°C), mechanically ground and sieved through a filter with a mesh to obtain <2 mm fraction. A 20–30 g-sub-sample was drawn from the bulk soil fraction, and re-ground to obtain <200 μm fraction using a mortar and a pestle. From this fine material, 1.0 g of soil sample was transferred into a digestion tube for digestion followed by analysis according to the USEPA method 3050B and data obtained treated as explained above. The soil samples were analyzed in triplicates and arranged in a similar way as the moss samples for data analysis.

Results and Discussion

The characteristics of the soil samples taken from the sampling points were determined and it was found that soil samples from Webuye area had a loam texture, a pH of 5, cation-exchange capacity of 12.8 meq/100 g, 1.9% organic carbon, 56.8% sand, 32% silt and 11.2% clay. The rainfall pattern at Webuye and at control site (Kakamega) between April and June, prior to and during the wet season sampling, was similar and was recorded as: at Webuye the first month mean of 82.5 mm (weekly range 70–140 mm), the second month mean of 52.5 mm (weekly range 25–75 mm), the third month mean of 44 (weekly range 10–60 mm), at Kakamega the first month mean of 111.3 mm (weekly range 30–245 mm), the second month mean of 45 (weekly range 25–60 mm) and the third month mean of 51.3 mm (range 25–70 mm). There was negligible rainfall during the second sampling in January 2007 in both Webuye and Kakamega. The analytical method found mean % recovery values for Cr, Pb, Cu, Zn and Fe of 93, 89, 94, 88 and 84 for both soil and moss, with detection limits of 0.001, 0.001, 0.003, 0.008 and 0.01 ng mL^{-1}, respectively.

Table 1 contains the mean concentrations of the various metals in soil samples (in μg/g dry weight) at the different sites in Webuye, as well as the standard deviations from the means, irrespective of the seasons. The data in this table were obtained by combining all the metal concentration results in samples taken in each season in the south, west, east and north directions i.e. 100 m, 500 m and 1 km from factory, respectively. These data would indicate if there were any variations in the mean concentrations in samples taken within 1 km radius from the factory and the influence of the wind. In this study, it was considered that heavy metal levels in the topsoil would be determined by the geology, atmospheric deposition and redistribution due to leaching and runoff during rainfall. Elevated levels in comparison with control would indicate anthropogenic atmospheric deposition. Due to preferential flow of effluent runoff through canals and eventual deposition in water and sediment of river Nzoia, effluent from point sources such as the Pan Paper factory and the nearby East African Heavy Chemical industry, would mainly impact on water canals and river Nzoia. Topsoil concentrations would therefore mainly indicate an influence of atmospheric deposition.

It is evident from Table 1 that copper is evenly distributed on all sides of the industry, with the southern side and the eastern side having slightly higher concentrations. Zinc was more concentrated on the south, the western and the eastern sides and was slightly in higher amounts compared with all the rest of the metals except iron. Iron was also evenly distributed in all directions and had the highest concentrations. The differences in mean concentrations with respect to directions from the factory can be attributed mainly to wind direction, which directs the atmospheric emissions from the factory towards the south-western side of the factory and, possibly, to wash-off by rain from corrugated iron roofs in the east. Cadmium and lead showed relatively lower concentrations in comparison with all the other metals especially in the eastern and northern directions. However, samples from the south and

Table 1 Mean (±SD) concentrations (mg/kg dry weight) of heavy metals in soil samples based on groups of data at different sites, seasons and distances from the factory, respectively

Site	Cu	Zn	Cr	Cd	Fe	Pb
South	31.8 ± 0.3	57.2 ± 1.2	40.5 ± 4.6	14.8 ± 0.1	2,266 ± 13	38.5 ± 3.4
West	29.1 ± 1.8	57.9 ± 3.5	37.6 ± 3.5	14.3 ± 1.1	2,275 ± 8	31.4 ± 18.1
East	32.5 ± 5.2	78.9 ± 16.6	41.0 ± 7.7	14.1 ± 1.4	2,267 ± 4	9.2 ± 8.2
North	28.4 ± 2.7	42.7 ± 4.1	34.1 ± 3.9	14.0 ± 1.1	2,198 ± 77	9.9 ± 1.5
Wet season	29.5 ± 1.2	68.5 ± 7.7	34.8 ± 3.3	14.1 ± 0.2	2,235 ± 67	14.8 ± 1.2
Dry season	31.4 ± 0.4	49.8 ± 2.4	41.8 ± 4.4	14.4 ± 2.1	2,268 ± 12	29.7 ± 2.2
100 m	28.8 ± 2.2	62.2 ± 4.1	31.7 ± 6.4	14.2 ± 1.0	2,223 ± 10	38.2 ± 12.1
500 m	29.7 ± 3.1	54.1 ± 4.4	37.0 ± 3.3	14.0 ± 0.4	2,221 ± 14	7.4 ± 2.1
1 km	32.8 ± 1.1	61.2 ± 3.8	46.2 ± 1.2	14.4 ± 1.1	2,310 ± 11	20.5 ± 1.6

n = 24; Control soil from Kakamega contained mean metal concentrations (mg/g dry soil) of 75.8 (Fe), 6.6 (Cu), 13.2 (Zn), 8.8 (Cr), 3.4 (Cd) and 8.4 (Pb) from Muendo (2009) west sides of the factory showed significantly higher concentrations of lead which can be attrributed to the influence of the wind direction carrying factory emissions to these directions among other factors such as use of leaded fuel at the motor yard.

Iron (Fe) showed the highest concentration of all the heavy metals. This is mainly due to its geological abundance as Fe would be expected to occur at higher concentrations even in non-contaminated soils (Romic and Romic 2003). Earlier, analysis of the control soil sample from Kakamega gave a mean Fe-concentration of 75 mg/kg dry soil (Muendo 2009) indicating the natural background level of the region. From our results, the abundance trend of these heavy metals in the topsoils was thus Fe > Zn > Cr > Cu > Pb > Cd. Apart from emissions from the factrory, rusting due to the high acidity in the environment surrounding Pan African Paper Mill due to the presence of the nearby East African Heavy Chemical industry could also play a role in the bioaccumulation of Fe and Zn in the soils. The East African Heavy Chemical industry, located on the southern-western side of the Pan Paper factory, contributes to the rusting and corrosion observed in the area. However, washoff from the roofs would mostly go into the drainage channels during rainfall and finally end up in River Nzoia. Analysis of variance (ANOVA) did not show significant difference ($p < 0.05$) in the concentrations of all the heavy metals in the various directions of the industry, but there was significant difference with direction for Zn and Pb, respectively. For the topsoil samples, although the wind directions were expected to contribute significantly towards their heavy metal load variations, it was not possible to make this conclusion for all metals, indicating that other factors such as rainfall, corrosion of iron roofs and, to a limited extent, emissions from other industrial activities within the town were also possibly influencing the distribution of heavy metals in soil around the Pan Paper Mill.

Spearman's Correlation analysis was done on the metal concentrations in the soil samples collected around the factory (Table 2) and it was noted that Fe had reasonable significant correlation values with the elements Zn and Cr. There was also a significant ($p < 0.05$) positive correlation between Cu and Zn, Cu and Cr, Zn and Cr and between Cd and Pb. This indicated possible common sources of these pairs of metals and the common sources include aerial deposition from the factory. Aerial deposition from the factory and from other industrial activitiers in Webuye were therefore seen to have an influence on the concentration of these metals in the soils around the factory. The soil heavy metal concentration data were also combined to show contributions from seasonal variations. Table 1 also shows the mean concentrations of the heavy metals in soils in the two different seasons in mg/kg of dry weight. These data were generated by combining all 24 samples taken from around the factory, i.e. from a radius of 100 m, 500 m and 1 km, regardless of the direction. Generally, the mean metal concentrations of Cu, Cr, and Pb were higher during the dry season compared with the wet season, indicating aerial deposition as the likely source. However, the concentrations of Cd and Fe did not show any significant seasonal variation. The mean Zn concentration in the soil samples taken in the wet season was higher indicating that it is likely influenced by wash-off by rain or wet deposition.

The higher concentrations of Cu, Cr, and Pb in the drier season could be attributed to their leaching and distribution from the topsoil by runoff water during wet weather resulting in lower concentrations compared to dry weather concentrations. During sampling, the wet season experienced very high amounts of rain. It is evident that the top (0–15 cmm) layer of the soils collected had little of the metals Cu, Cr and Pb due to the runoffs and seepage as a result of the dilution effect of the rainy season**.** The data show that Fe and Cd concentrations did not depend on proximity to the Paper Mill as higher concentrations of these metals in soil, 1 km away from the factory, were possibly due to synergistic effects such as reactions by acidic gases released by the Paper Mill as well as the East African Heavy Chemical industries. Wind movement was most likely responsible for transfer of metals to longer distances from the factroy. Generally, the concentrations of all metals were not siginificantly affected by the distance from the factory. High wind speeds could also support aerial transport to locations far away from the factory before deposition occurs.

For the heavy metal loads in the mosses around the factory (Table 3) the concentrations of Cu, Zn, Cr, Cd, and Fe were significantly ($p < 0.05$) higher compared with the control samples, sampled approximately 30 km away from the factory. These results clearly indicate the influence of the pulp and paper factory as well as other industrial activities such as those at the East African Heavy chemical industries in Webuye town on aerial distribution and deposition of these toxic heavy metals. However, the Pb

Table 2 Pearson's correlation matrix for mean total metal concentrations in the soils

	Cu	Zn	Cr	Cd	Fe	Pb
Cu	1					
Zn	0.815478*	1				
Cr	0.948306*	0.840262*	1			
Cd	0.423633	−0.00892	0.528208	1		
Fe	0.598057	0.707243*	0.819659*	0.492418	1	
Pb	0.094617	−0.14717	0.315896	0.889526*	0.556965	1

* Correlation is significant at $p < 0.05$

Table 3 Mean (± SD) total concentrations of heavy metals in mosses (in mg/kg dry wt) in the different directions f the paper factory

	Cu	Zn	Cr	Cd	Fe	Pb
South	34.2 ± 8.9	124.5 ± 1.9	26.9 ± 9.3	14.7 ± 1.4	1,984 ± 180	10.6 ± 15.2
East	29.5 ± 16.7	60.9 ± 55.6	24.4 ± 7.3	14.6 ± 0.2	2,079 ± 8.	23.4 ± 11.8
West	27.9 ± 1.0	110.4 ± 1.9	30.3 ± 4.2	13.5 ± 0.3	2,020 ± 205	20.0 ± 9.9
North	22.6 ± 6.7	82.7 ± 35.6	23.2 ± 13.7	13.4 ± 0.2	1,822 ± 12	3.0 ± 2.1
Control	17.1 ± 0.4	28.36 ± 0.5	17.4 ± 2.6	9.0 ± 0.7	853 ± 4	14.8 ± 3.7

n = 24

concentrations were comparable with those in control samples which can be attributable to ubiquitous nature of Pb as a consequence of other non-point sources such as emissions from vehicles. Leaded petrol, which could be a non-point source of Pb, was phased out in in Kenya in December 2006, just before sampling was done in 2007. Activities within the factory that could discharge toxic metals into the atmosphere include welding, painting, scrap metal fabrications, vehicular emissions and burning of fuel oil, biomass fuels and other waste products within the factory (Ruhling 1994, Singh 2001).

The mean concentrations of heavy metals Cu, Zn, Cr, Cd and Fe in mosses around Webuye Pan Paper factory were found to be significantly ($p < 0.05$) higher than in controls taken from Kakamega indicating atmospheric contamination mainly from the anthropogenic activities within the factory, deposition and accumulation by moss within a circumference of 1 km radius from the factory.

Figure 1 shows the mean concentrations of heavy metals in samples of moss in relation to distance from the factory. The pattern exhibited by heavy metal concentrations in mosses as shown in Fig. 1 is a clear indication of atmospheric depositions. For instance, Fe, Cu, and Cr concentrations were highest at 500 m. The particulate size and mass of heavy metal complexes are determined by both the atomic size, the electronic structure of the metal and the type of organic matter ligands binding the elements (Singh 2001, Krishna et al. 2003). Although the atomic sizes of these metals increase from Cr < Fe < Cu < Zn < Cd < Pb, the emissions that went farther than 500 m away from the factory seemed to carry along with them more of the heavier metals like Zn, Cd and Pb. ANOVA analysis showed no significant difference with distance possibly because the distances chosen in this study were relatively too close to one another and to the factory to depict any significant differences in heavy metal loads.

Figure 2 compares the mean concentrations of the heavy metals in mosses in different seasons. The data are obtained by combining all samples taken, separately, in two seasons. The mean concentrations of heavy metals Cu, Zn, Cr, Cd, Fe and Pb were higher in the dry season than wet season, which confirms that they are absorbed by the mosses mainly from atmospheric deposition. Similar temporal and site trends in heavy metal concentrations in moss as bio-indicators of atmospheric heavy metal pollution have been reported in other countries (UNECE 2008).

Different moss species have been used worldwide to monitor heavy metal pollution as a result of urbanization,

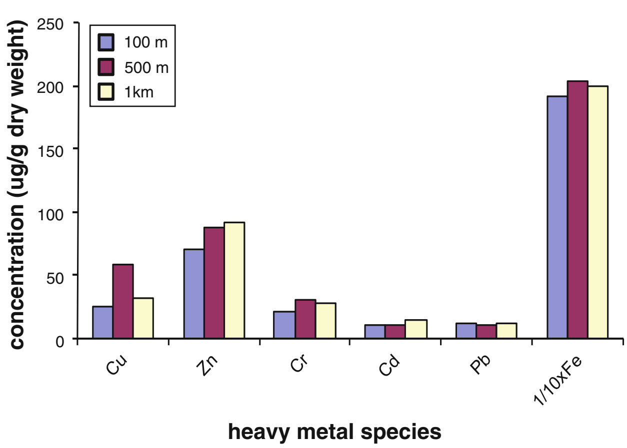

Fig. 1 The variation of mean concentrations of heavy metals (µg/g dry weight) in moss with distance from factory. (The actual mean concentrations of Fe are reduced by a factor of 1/10 before making the plots)

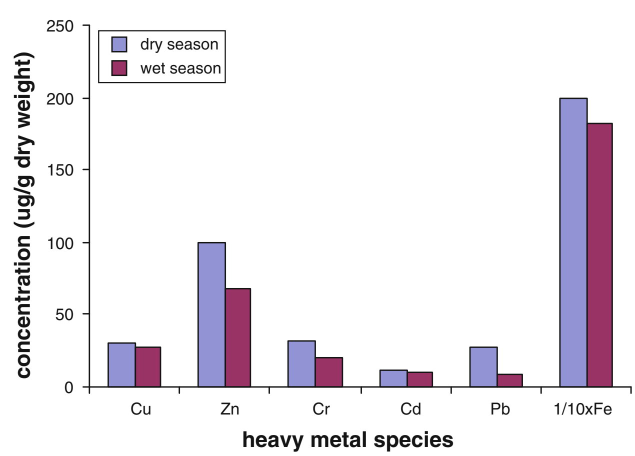

Fig. 2 The mean concentrations of heavy metals (µg/g dry weight) in moss showing seasonal variation. (The actual mean concentrations of Fe were reduced by a factor of 1/10 before making the plots)

Table 4 Comparison of mean concentrations (μg/g dry matter) of heavy metals in mosses and soil samples obtained from this study with those of other countries

Location	Zn	Cu	Fe	Cd	Co	Ni	Cr	Pb	Reference
Moss									
Webuye (Kenya)	60.9–124.5	22.6–34.2	1,822.4–2,079.2	13.3–14.7	NA	NA	20.1–33.3	3.0–26.9	This study
Istanbul	74.8	22.2	3,000	0.51	2.42	6.3	NA	NA	Belivermis et al. (2008)
Thrace	30.9	7.8	5,967	0.19	4.1	6.0	NA	NA	Coskun et al. (2006)
Bulgaria	32.6	14.5	1,410	NA	NA	3.1	NA	NA	Buse et al. (2003)
Romania	79.5	21.5	2,510	NA	NA	3.4	NA	NA	Buse et al. (2003)
Russia	34.9	6.5	616	NA	NA	2.3	NA	NA	Buse et al. (2003)
Galicia (Spain)	59.2	6.2	476	NA	0.5	1.6	NA	NA	Fernandez and Carballiera (2000)
India*	48.9	NA	2,760	0.73	NA	NA	NA	89.7	Saxena et al. (2008)
Soil									
Webuye (Kenya)	42.7–78.9	28.4–32.8	2,198.0–2,310.3	14.0–14.8	NA	NA	31.7–46.2	7.4–38.5	This study
Thrace (urban&rural)	45	20	26,900	0.2	11	5.4	NA	NA	Coskun et al. (2006)
Bangkok (urban)	118	41.7	16,100	0.29	NA	24	NA	NA	Wilcke et al. (1998)
Zagreb (urban)	77.9	20.8	27,000	0.69	NA	49	NA	NA	Romic and Romic (2003)
Galicia (urban&rural)	42.4	19.3	24,451	NA	13	11	NA	NA	Fernandez and Carballiera (2000)
Istanbul (urban)	82	28.7	9,635	0.14	8.2	23	NA	NA	Belivermis et al. (2008)

NA not analysed; * *Barbula constricta* sp. sample of 1999

industrialization and other anthropogenic activities and some of the data obtained from various references are compared with our data in Table 4. Although it is not possible to fully justify any comparison of our data with these Asian and European data because of differences in climate and the levels and types of industrial activities, this comparison provides a scientific basis for the use of indigenous moss species as biomonitors of atmospheric heavy metal pollution worldwide.

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