Heavy Metal and Radioactivity Concentrations in Soil and Moss Samples from Istanbul, Turkey

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Abstract

Concentrations of Cd, Cu, Co, Cr, Fe, Ni and Zn in the moss (*Hypnum cupressiforme* and *Scleropodium purum)* and topsoil (0-5 cm) samples collected from Istanbul were analysed to estimate heavy metal deposition. The activity concentrations of gamma emitting radionuclides (^{137}Cs, ^{40}K, ^{238}U, ^{232}Th), organic matter contents and pH values of the topsoil samples were also measured. The trace element and radionuclide levels were compared with those found in the samples from other locations of Turkey and different countries.

Keywords: Heavy metal, radioactivity, moss, soil, Istanbul.
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Introduction

The using of mosses as biomonitors of airborne heavy metal depositions, was firstly used at the end of the 1960s (Rühling and Tyler 1968) and has been widely used for the last four decades (Mishev et al. 1996; Rühling and Steinnes 1998; Gerdol et al. 2000). Mosses have several advantages comparing to higher plants in these studies. Foremost, they do not have root system, therefore they supply their nutrient needs from absorbtion of atmospheric particles deposited by dry or wet on their surfaces (Berg and Steinnes 1997a; Rühling and Steinnes 1998; Szczepaniak and Biziuk 2003). Thus, they accumulate atmospheric trace elements and retain them for long periods of time. Mosses have also large surface/mass ratio. It has been estimated that mosses have approximately 10 times more large surface area compared to that of herbaceous grasses (Mishev et al. 1996). Eventually, because of their high surface/volume ratio and absence of cuticle and epidermis, they accumulate trace elements, concentrating them in tissues (Adamo et al. 2003). Consequently, using mosses as biomonitors of atmospheric pollution is quite useful method to determine sort, quantity and sources of atmospheric pollutants (Steinnes 1995).

Soil is the main reservoir for pollutant elements and the principal source of heavy metals and radionuclides entering the food chain. Organic and inorganic material contents of topsoil depend on mainly physicochemical properties of soil, vegetation type, climatic conditions and extent of anthropogenic effects (Niskavaara et al. 1997). The analysis of topsoils, has been widely used to evaluate the uptake of contaminants in ecosystems (Kabata-Pendias and Pendias 1984; Lee and Lee 1997; Niskavaara et al. 1997).

It is known that most of the radioactivity that humans are exposed arises mainly from natural radionuclides. The three main components of natural radiation (^{232}Th and ^{238}U series and ^{40}K) typically deliver an approximately equal level of external gamma radiation to individuals both outdoors and indoors (UNSCEAR 2000). It is believed that ^{137}Cs in a soil deposited by the atmospheric weapon tests and nuclear power plant accidents

and is remained in biota and living organisms because of its relatively long physical half-life (Lee and Lee 1997).

The aims of this study are: (1) To determine the levels of heavy metal in moss and topsoil samples, and gamma emitting radionuclides (^{137}Cs, ^{40}K, ^{238}U, ^{232}Th) in the topsoil samples (2) to determine annual variations in the heavy metal concentrations (3) to learn the relationships among heavy metal content, pH and organic matter of soil samples.

Material and Methods

Study area and sampling procedure

The sampling site Istanbul metropolis is located in the northwest part of Turkey. Istanbul has about 15 million inhabitants and is the most industrialized and largest city in Turkey. The arithmetic mean of annual rainfall in Istanbul is in the range of 650-900 mm and northern winds are dominant.

The samples were collected from 7 sites in December of 2004 and 2006 years. Coordinates of all sampling stations were recorded using a GPS (Global Positioning System). Each sampling site was chosen to be at least 150 m away from the main roads and populated areas and at least 50 m away from secondary roads or houses. Geographical Information System (GIS) technique was used for constructing the map of the study area. The study area and the sampling points were shown in Figure 1.

In forests and plantations, moss and topsoil samples were collected from small open areas to prevent canopy and dripping effects of trees. The moss samples carefully cleaned from soil and other debris, and they were not washed prior to drying. The topsoil samples were collected from flat and undisturbed sites. In the laboratory, the soil samples were dried at 40 ^{0}C to a constant weight, plants and stones were removed, and the remaining soil passed through a 2 mm sieve and homogenized.

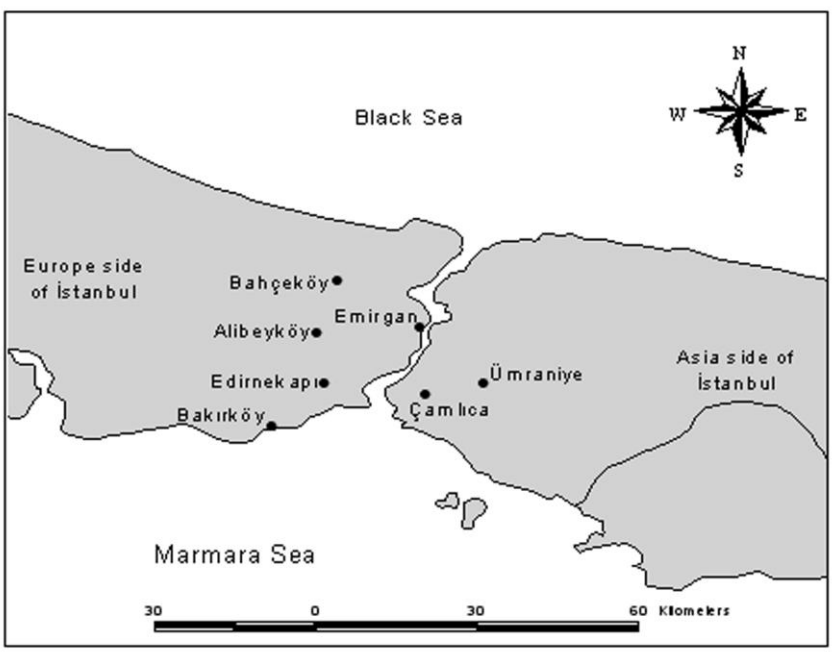

Figure 1. Study area and sampling stations

Analytical methods

The organic matter contents of the soil samples were determined at 550 ^{0}C for 4 h using the "loss-on-ignition" method (Allen 1989). The pH was measured in suspension of soil in bi-distilled water. For this purpose, 10 gr of soil sample mixed up in 25 ml bi-distilled water for 3 minutes, and 30 minutes afterwards pH measured by means of pH-meter. In order to remove organic substances, glass and plastic equipments which were used in chemical treatments, had been held in 6 M HNO_3 solution for two days, afterwards rinsed with de-ionized water for six times.

For digestion, 1 gr of the moss and soil was wet ashed. The sample was put into a glass beaker, 5 ml concentrated H_2SO_4 added and heated on the hot plate up to 90 ^{0}C. Two hours afterwards, a few drops concentrated HNO_3 was added very slowly and continued heating at 120 ^{0}C. When the sample solution becomes liquid hydrogen peroxide was added and heating was continued at 120 ^{0}C for 30 min. The hydrogen peroxide was added until the sample remained clear for two hours at 150 ^{0}C. After that, the sample was filtered and diluted to 20 ml with 2 % HNO_3 (Chapman 1992). These final solutions were analysed for Zn, Cd, Cu, Fe, Co, Ni, and Cr using atomic absorption spectrophotometry (Shimadzu AA 680, air–C_2H_2, background correction mode). The concentrations in each sample solution measured for three times. Blank solutions were prepared under the same conditions as the samples to control possible contamination during the preparation procedures of samples. Accuracy of the digestion and measuring processes checked by analysis of reference material from MAPEP (Mixed Analyte Performance Evaluation Program).

For radionuclide analysis, soil samples were placed into 1-liter Marinelli type beakers and radiation levels were measured using a gamma multichannel analyser equipped with an HPGe detector. The gamma spectra were analyzed using the ORTEC Maestro 32 data acquisition and analysis system. The detector had coaxial closed fasing geometry with following specifications: Resolution (FWHM) at 122 keV ^{57}Co is 1.0 keV and at 1.33 MeV ^{60}Co is 2.0 keV. Relative efficiency at 1.33 MeV ^{60}Co is 22.1%. The detector was shielded by a cylindrical lead shield with an average thickness of 10 cm, in order to achieve the lowest background level. The energy dependent efficiency calibration was done using a standard soil nuclide mixture (Isotop Products Laboratories) containing known activities of ^{241}Am, ^{57}Co, ^{60}Co, ^{134}Cs, ^{137}Cs and ^{54}Mn. Decay periods were 24 hours for short-lived and 3-4 weeks for long-lived isotopes. The activity concentration of ^{40}K was determined from the peak areas at 1460 keV. The 661.6 gamma transition was used to calculate the ^{137}Cs activity concentration. The activity concentrations of ^{238}U and ^{232}Th were calculated assuming secular equilibrium with their decay products. The gamma transition lines of ^{226}Ra (186.1 keV), ^{214}Pb (351.9 keV) and ^{214}Bi (609.2 keV) were used to calculate specific activities of radioisotopes in the U-series. The spesific activities of radioisotopes in the Th-series were determined using the gamma transition lines of ^{212}Pb (238.6 keV), ^{208}Tl (583.1 keV), ^{228}Ac (911 keV). The mean counting time for each sample was 10 hours.

Results

Heavy metal concentrations of the moss and soil samples were presented in Table 1 and Table 2, respectively. Heavy metal levels of moss and soil samples from various studies were also presented in Table 3 and 4, respectively.

Heavy metal concentrations found in Istanbul were generally higher compared with those found in the neighbour regions except for the findings of Eğili et al. listed in Table 3. However, the heavy metal levels of topsoil samples were similar to those found in different studies both urban and rural sites (Table 4). The activity concentrations of ^{40}K, ^{232}Th and ^{238}U in the soil samples were lower than the world reference values (UNSCEAR 2000).

Table 1. Heavy metal concentrations in moss samples (μg g^{-1})

Sampling stations	Year	Zn	Cd	Cu	Fe	Co	Ni	Cr
Alibeyköy	2004	72.6	0.61	23.1	2.9**	2.39	6.4	9.6
	2006	47.7	0.41	14.3	3.6	2.49	3.3	6.4
Bahçeköy	2004	80.2	0.41	14.7	3.5	2.28	7.1	16.9
	2006	57.9	0.49	14.4	1.8	2.90	4.7	7.0
Ümraniye	2004	90.7	0.60	24.8	2.5	1.40	5.1	8.3
	2006	69.5	0.76	20.9	1.3	1.86	3.3	4.9
Çamlıca	2004	72.2	0.69	18.1	1.6	1.50	3.2	8.5
	2006	78.2	0.58	13.2	3.5	2.61	3.6	13.1
Edirnekapı	2004	144.7	0.67	62.0	2.9	2.35	12.2	14.7
	2006	86.3	0.58	28.9	2.0	1.81	4.9	7.9
Bakırköy	2004	97.2	0.21	26.5	4.1	3.11	11.7	7.0
	2006	101.0	0.32	36.9	2.9	2.88	9.5	10.4
Emirgan	2004	77.9	0.36	21.2	4.5	<0.5	10.5	13.4
	2006	83.1	0.41	26.6	6.0	<0.5	14.9	20.4
Arithmetic mean	2004	90.8 ± 25.5 *	0.51± 0.18	27.2 ± 15.9	3.1 ± 1.0	2.17 ± 0.64	8.0 ± 3.5	11.2 ± 3.8
	2006	74.8 ± 18.0	0.51 ± 0.15	22.2 ± 9.0	3.0 ± 1.6	2.42 ± 0.48	6.3 ± 4.4	10.0 ± 5.3

* standard deviation ** mg g^{-1}

Table 2. Heavy metal concentrations in soil samples (µg g^{-1})

Sampling stations	Year	% organic matter	pH	Zn	Cd	Cu	Fe	Co	Ni	Cr
Alibeyköy	2004	18.5	5.5	100.1	0.22	32.6	11.3**	13.2	30.3	43.5
	2006	19.0	5.5	105.5	0.48	33.0	11.1	12.8	30.6	43.1
Bahçeköy	2004	11.5	6.7	136.2	n.d	28.0	10.0	9.3	22.8	41.1
	2006	14.0	6.0	73.5	n.d	17.8	10.0	8.1	21.5	43.5
Ümraniye	2004	7.5	6.7	83.9	0.24	13.2	8.7	3.4	13.0	24.3
	2006	7.0	6.6	83.1	0.18	30.5	7.8	3.4	9.7	24.4
Çamlıca	2004	8.0	5.5	42.0	0.12	29.4	9.8	5.9	13.2	32.8
	2006	7.5	5.3	33.8	0.09	27.8	9.7	5.4	14.4	34.7
Edirnekapı	2004	10.5	7.7	91.4	0.09	43.9	9.7	11.1	55.9	75.0
	2006	10.5	7.8	88.5	0.02	31.8	9.4	10.2	37.9	46.5
Bakırköy	2004	9.5	7.7	83.4	0.02	21.0	8.2	4.0	18.3	25.5
	2006	17.0	7.7	104.8	0.03	27.5	8.2	3.9	20.4	22.0
Emirgan	2004	12.0	6.6	86.6	0.05	24.6	10.8	18.9	35.7	50.4
	2006	9.5	6.6	85.1	0.05	32.8	11.2	13.5	33.2	35.0
Arithmetic mean	2004	11.1 ± 3.7*	6.6 ± 0.9	89.1 ± 27.8	0.12 ± 0.09	27.5 ± 9.6	9.8 ± 1.1	9.4 ± 5.6	27.0 ± 15.3	41.8 ± 17.5
	2006	12.1 ± 4.7	6.5 ± 1.0	82.0 ± 24.2	0.14 ± 0.18	28.7 ± 5.3	9.6 ± 1.3	8.2 ± 4.1	23.9 ± 10.3	35.6 ± 9.6

* standard deviation ** mg g^{-1}

Table 3. Heavy metal concentrations in moss samples from various studies (µg g^{-1})

Reference	Area	Zn	Cu	Fe	Cd	Co	Ni	Cr
Coşkun et al. 2005	Thrace	30.9	7.8		0.19		6.6	
Eğili et al. 2003	Thrace	252.7		5967		4.1		17.6
Buse et al. 2003	Bulgaria	32.6	14.5	1410			3.33	2.4
Buse et al. 2003	Romania	79.5	21.5	2510			3.35	8.5
Buse et al. 2003	Russia	34.9	6.5	616			2.0	1.5
Fernandez and Carballeira 2000	Galicia (Spain)	59.2	6.2	476		0.5	1.8	1.4
Present study	Istanbul	74.8	22.2	3000	0.51	2.42	6.3	10.0

Table 4. Heavy metal concentrations in topsoil (0-5 cm) samples from various studies (μg g^{-1})

Reference	Area	Zn	Cu	Fe	Cd	Co	Ni	Cr
Coşkun et al. 2006	Thrace (urban and rural)	45	20	26900	0.2	11	50	173
Wilcke et al. 1998	Bangkok (urban)	118	41.7	16100	0.29		24.8	26.4
Romic and Romic 2003	Zagreb (urban)	77.9	20.8	27000	0.66		49.5	
Fernandez and Carballeira 2000	Galicia (urban and rural)	42.4	19.3	24451		13	11.1	100.2
Present study	Istanbul (urban)	82.0	28.7	9635	0.14	8.2	23.9	35.6

Table 5. The activity concentrations of ^{137}Cs, ^{40}K, ^{238}U and ^{232}Th in soil samples (Bq kg^{-1})

Sampling stations	^{137}Cs	^{40}K	^{238}U	^{232}Th
Alibeyköy	27.0 ± 0.37*	578.6 ± 12.4*	19.5 ± 1.20*	20.5 ± 1.86*
Çamlıca	11.7 ± 0.30	242.7 ± 6.3	25.9 ± 1.85	23.5 ± 2.42
Emirgan	17.3 ± 0.28	311.3 ± 7.0	16.6 ± 1.13	20.4 ± 1.66
Edirnekapı	6.9 ± 0.26	532.6 ± 11.7	28.2 ± 1.62	36.1 ± 1.39
Bakırköy	29.4 ± 0.48	302.2 ± 7.7	11.4 ± 1.27	13.6 ± 2.07
Ümraniye	19.2 ± 0.35	156.9 ± 4.4	19.4 ± 1.67	17.6 ± 1.97
Bahçeköy	7.5 ± 0.21	285.7 ± 6.7	19.8 ± 1.31	20.4 ± 2.09
Arithmetic mean	17.0 ± 8.9**	344.3 ± 153.8	20.1 ± 5.6	21.7 ± 7.1

* Standard deviation of measuring ** Standard deviation of arithmetic mean

Table 6. ^{40}K, ^{238}U and ^{232}Th activity concentrations (Bq kg^{-1}) in various studies

Reference	Area	^{40}K	^{238}U	^{232}Th
UNSCEAR 2000	worldwide	400	35	30
Karahan and Bayülken 2000	Istanbul	342	21	37
M.S. Al-Masri et al. 2006	Syria	336 ± 116	19 ± 10	24 ± 10
Ramzaev et al. 2006	Russia	520		30
Present study	Istanbul	344.3 ± 153.8 *	20.1 ± 5.6	21.7 ± 7.1

* Standard deviation of arithmetic mean

Table 7. Pearson's correlation coefficients among %organic matter, pH and heavy metals

	Zn	Cd	Cu	Fe	Co	Ni	Cr	^{137}Cs
% organic matter	0.52	0.35	0.18	0.45	0.41	0.35	0.29	0.56
pH	0.39	-0.49	0.07	-0.52	-0.14	0.35	0.08	0.20

In general, weak positive correlations were observed between heavy metal and organic matter contents in soil samples.

Discussion

Heavy metal and radioactivity concentrations

In general, study areas contain both rural and urban regions in the Table 3 except for present study. The comparatively high heavy metal values can be attributed to the sampling sites near to urban areas in this study. It was observed that average Co, Cr, Fe and Ni concentrations of the soil samples were higher than that of the moss samples, similar to findings of Fernandez and Carballeira (2000). On the other hand, Zn and Cu contents were similar, Cd concentrations were pretty higher in moss samples comparatively to the soil samples. It may be clarified with that Cu, Pb, Cd and Zn accumulations in moss are more dynamic process comparatively to those of soil. While the heavy metal present of moss reflects approximately last three years accumulation, heavy metal accumulation in soil is a long term process. Consequently, heavy metal inventory of moss samples depend on the dry and wet deposition originated by local sources, long range transport and enrichment by soil approximately for last three years. Furthermore, it was early shown that Cu, Pb, Cd and Zn derive mainly from human activities such as metal industry and fossil fuel combustion (Vernet 1991) and they are well identified as air pollution markers in several investigations (Loppi et al. 1997; Bargagli et al. 2002). These elements can be transported long range in atmosphere and accumulate in unpolluted areas by precipitation rather than soil enrichment (Pacyna et al. 2001).

The previous radioactivity concentration values reported by Karahan and Bayulken (2000) for Istanbul were very similar to those found in this study except for ^{232}Th (Table 6). It is known that northern hemisphere has exposured more fall-out radionuclides than southern hemispere as a result of the abundance of the nuclear activities in the northern hemisphere. Therefore, it was advisable to compare activity concentration of ^{137}Cs found in this study with other countries which are in northern hemisphere (Table 8). In this study, the activity concentration values of ^{137}Cs were lower than the reported values from the countries in the northern hemisphere.

Table 8. ^{137}Cs activity concentrations in 5 cm depth soil in various studies

Reference	Area	Mean ± standard deviation (Bq kg^{-1})	Range
Topçuoğlu et al. 2003	Northeast part of Turkey		7 – 52
Kim et al. 1998	South Kore	33.2 ± 16.1	7.86 - 70.1
Elejalde et al. 1996	Spain	29.7 ± 16.0	
Present study	Istanbul	17.0 ± 8.9	6.9 – 29.4

Annual variations

It was not observed important differences between average heavy metal concentrations

measured in 2004 and 2006 years (t test, $p>0.05$). However, variations were observed among the sampling points for the moss samples. For instance, while Cd and Cu contents in moss samples decreased from the years 2004 to 2006 in Çamlıca sampling station, the same contents increased in Bakırköy station between these years.

Fe, Co, Ni and Cr concentrations in the soil samples in 2004 and 2006 were very similar. It was shown that these elements were mainly natural in origin (Coşkun et al. 2006). For this reason, atmospheric long range transport of these elements has poor effect on soil concentrations. In a previous study, Cr was found highly bound in soil and is of little importance in other sources (Berg et al. 1996). The noticeable temporal variations of the elements originating mainly from human activities in the surface soil may be explained with the fluctuations of extent of anthropogenic effects rather than the physico-chemical properties of soil such as organic matter content, pH value and cation exchange capacity.

It was reported that Zn, Cd, Pb and Hg concentrations in moss has decreased in recent years owing to the fact that West Europe countries restricted of elements dispersing from their industry (Berg and Steinnes 1997). Meanwhile, it was not reported considerable temporal variations in Cu, Ni and Co concentrations of moss (Berg and Steinnes 1997).

Effect of organic matter and pH

Soil organic material is originated from vegetation and animal sources, and is one of the major factors controlling the physical and chemical properties of soil including the metal bounding capacity, buffering capacity and water holding capacity. Thus, it affects deposition, vertical and horizontal migration of heavy metals and radionuclides in soil. Other major physicochemical factors affecting the retention of these pollutants in soil are clay minerals, secondary hydroxy and oxide precipitates (Niskavaara et al. 1997). Besides, in the urban regions, which are normally comprise major roads, industrial plants and inhabiting areas, physico-chemical properties of the soil such as the organic matter content have a limited impact on the retention atmospheric heavy metals. Since, multiplicities and properties of local heavy metal sources assign deposition patterns in urban areas. Conversely, in the rural areas, because of the fact that the rural areas normally have not local pollution sources, heavy metals reach to the surface of the soil through atmospheric long range transport, accumulate in the topsoil and finally distribute vertically and horizontally in respect to physico-chemical properties of the soil.

It is well known that soil organic matter is the main factor affecting ^{137}Cs deposition in surface soil (Elejalde et al. 1996; Lee and Lee 1997). In this study, correlation coefficient between organic matter content and ^{137}Cs in the surface soil was found to be 0.56 (Table 8). The higher correlation coefficient might have been observed if the ^{137}Cs amounts were measured vertically in each sampling station.

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