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Radioactive Contamination in Lichens Collected from Trabzon and Rize in the Eastern Black Sea Region, Turkey, and a Comparison with that of 1995

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Abstract After the Chernobyl nuclear accident in 1986, 14 lichen specimens collected from Trabzon and Rize in 1995 were used to detect the amount of the fallout radionuclides. In this paper, radioactivity levels in the same species from the same localities were re-detected to compare with those of 1995. According to the results of the two studies, the radioactivity levels that this paper found for ^{137}Cs and ^{40}K are significantly lower than those of 1995 (about 5.5–127 folds for ^{137}Cs, 5–17 folds for ^{40}K). The level of ^{212}Pb was acceptably small. The highest activities of the lichen species are seen in the locality of Helvacı (Trabzon, 100 m) and Çamlıhemşin (Rize, 1850 m) while the activities are medium in Bozdoğan (Trabzon, 150 m), Kemaliye (Trabzon, 750 m) and Çamlihemşin (Rize, 900 m). The activity values generally increase significantly depending on the altitudes. Since the measurements were performed 20 years after the Chernobyl accident in 1986, radionuclides of ^{134}Cs having short half-life (2.062 y) have not been detected. All data was obtained with a coaxial high purity Ge detector of 15% relative efficiency and resolution 1.9 keV at the 1332 keV gamma of ^{60}Co (Canberra, GC 1519 model).

Keywords Chernobyl · Fallout · 137Cs · Turkey

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Numerous studies have been published on the radioactive contamination of lichens in European countries after the Chernobyl accident (Paatero et al. 2007; Pipiska et al. 2007; Puhakainen et al. 2007). Additionally, the main fallout radionuclides after Chernobyl accident was detected in moss, reindeers and also tea plants in Europe (LaBrecque and Cordoves 2007; Skuterud et al. 2005; Topcuoglu et al. 1997). In some studies, lichens have been used in monitoring surveys for radioactive pollution (Golubev et al. 2005; McClenahen et al. 2007; Rossbach and Lambrecht 2006). In Turkey, the observations for radioactive contamination were mostly performed in the Eastern Black Sea Region after the Chernobyl accident (Baysal and Yazıcı 1995; Gokmen et al. 1996; Köse et al. 1994; Saka et al. 1997). In past few years in the Black Sea region of Turkey, such studies have not been carried out by using lichens. In the spring–summer period of 2001, the soil-to-grass transfer of ^{137}Cs and also its relation to several soil properties were studied in Serbia (Krstic et al. 2007).

In this paper, the levels of potassium, lead and cesium radionuclides in 12 lichen specimens collected from Trabzon and Rize in the eastern Black Sea region of Turkey were determined 20 years after the Chernobyl accident in 1986. Additionally, the data were compared with those of 1995.

Materials and Methods

After the Chernobyl accident in 1986, 14 lichen samples that were collected from Trabzon and Rize in 1995 were used to detect the radioactivity levels (Baysal and Yazıcı 1995). In this work, except for sample 8 (*Xanthoria*

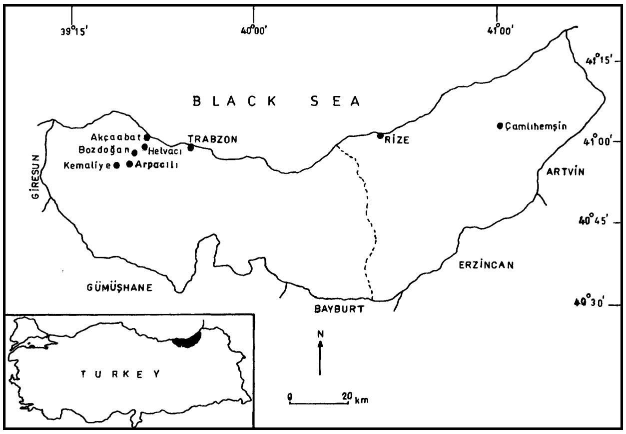

Fig. 1 A map showing the collecting sites

parietina) and sample 14 (*Lobaria pulmonaria*), the radioactivity levels in the same species collected from the same localities (Trabzon and Rize) in 2006 were re-detected to compare with those of 1995. The samples were collected from Akçaabat (Trabzon) and Çamlıhemşin (Rize) in the eastern Black Sea region of Turkey in 2006. Figure 1 shows a detailed description of all sampling sites. The soil samples were crushed thoroughly and dried at room temperature to a constant weight and later were crushed to pass through ca 2 mm mesh sieve to homogenize them.

After lichen samples (*Cladonia rangiformis*, *C. foliacea*, *C. furcata*, *C. portentosa*, *Dermatocarpon miniatum*, *Diploschistes ocellatus*, *Flavoparmelia caperata*, *Peltigera praetextata*, *Xanthoria parietina* and *Xanthoparmelia conspersa*) were separated from their substrata and were dried in an oven at 105°C for 24 h to ensure that moisture is completely removed and then were powdered. About 100 g of sample material were used and were sealed in gas tight, radon impermeable, cylindrical polyethylene plastic containers (5.5 cm diameter and 5 cm height) for gamma activity analysis. Each sample was sealed for 30 days to reach radioactive equilibrium where the decay rate of the daughters becomes equal to that of the parent. The locations, the altitudes and lichen species are given in Table 1. The radioactivity levels for lichens are given in Table 2.

Gamma spectrometry measurements were done with a coaxial high purity Ge detector of 15% relative efficiency and resolution 1.9 keV at the 1,332 keV gamma of ^{60}Co (Canberra, GC 1519 model). The detector was shielded within a 10 cm thick lead well internally lined with 2 mm Cu foils. The detector output was connected to a spectroscopy amplifier (Canberra, Model 2025). The energy calibration and relative efficiency calibration of the spectrometer were carried out using calibration sources which contained ^{133}Ba, ^{57}Co, ^{22}Na, ^{137}Cs, ^{54}Mn, and ^{60}Co peaks for energy range between 80 and 1,400 keV. The counting time for each sample and background was 50.000 s (Çevik et al. 2008).

The gamma ray transitions of energies 295.2, 661.6 and 1,460.8 keV were used to determine the concentration of the ^{212}Pb, ^{137}Cs and ^{40}K, respectively. The activity concentrations for the natural radionuclides in the measured samples were computed using the following relation

$$C_s = \frac{N_a}{\varepsilon P t w}$$

where C_s is in (Bq kg^{-1}), N_a is the net counting rate of gamma ray, ε is the counting efficiency of the used detector, *P* the absolute transition of gama decay, *t* the

Table 1 Locations, altitudes and species for the lichen specimens

Sample no.	Species	Locality	Altitude (m)
1	*Cladonia furcata*	Akçaabat-Bozdoğan-TRABZON	150
2	*Cladonia portentosa*	Akçaabat-Bozdoğan-TRABZON	150
3	*Cladonia rangiformis*	Akçaabat-Bozdoğan-TRABZON	150
4	*Xanthoparmelia conspersa* (Syn. *Parmelia conspersa*)	Akçaabat-Bozdoğan-TRABZON	150
5	*Parmotrema perlatum* (Syn. *Parmelia perlata*)	Çamlihemşin-RİZE	1,850
6	*Flavoparmelia caperata* (Syn. *Parmelia caperata*)	Çamlihemşin-RİZE	900
7	*Flavoparmelia caperata*	Akçaabat-Kemaliye-TRABZON	750
8	*Xanthoria parietina*	–	–
9	*Xanthoria parietina*	Akçaabat-Helvaci-TRABZON	100
10	*Dermatocarpon miniatum*	Akçaabat-Helvaci-TRABZON	100
11	*Diploschistes ocellatus*	Akçaabat-Helvaci-TRABZON	100
12	*Peltigera praetextata*	Akçabat-Arpacili-TRABZON	400
13	*Peltigera praetextata*	Çamlihemşin-RİZE	550
14	*Lobaria pulmonaria*	–	–

"–": not collected

Table 2 Radioactivity levels of lichens (Bq kg^{-1} dry weight)

Sample no	(Bq kg^{-1} dry weight)			
	^{212}Pb	^{134}Cs	^{137}Cs	^{40}K
1	6.75 ± 1.62	nd	24.098 ± 8.71	DL
2	18.90 ± 1.37	nd	53.66 ± 9.27	DL
3	10.35 ± 2.74	nd	28.94 ± 7.01	249.15 ± 21.00
4	17.16 ± 0.89	nd	152.78 ± 10.69	164.4 ± 10.61
5	35.34 ± 0.56	nd	438.24 ± 17.18	591.54 ± 24.27
6	32.79 ± 1.89	nd	179.99 ± 16.71	154.13 ± 11.82
7	28.49 ± 1.84	nd	196.54 ± 15.32	DL
8	–	–	–	–
9	24.43 ± 0.75	nd	22.65 ± 5.49	398.76 ± 11.52
10	12.89 ± 1.39	nd	73.12 ± 10.42	DL
11	17.18 ± 1.22	nd	429.53 ± 14.37	205.12 ± 10.27
12	20.29 ± 0.14	nd	31.57 ± 9.26	361.32 ± 23.27
13	7.11 ± 1.63	nd	40.59 ± 9.74	200.32 ± 11.52
14	–	–	–	–

"nd": not detected, "–": not collected and unmeasured, DL: detection limit

counting time in seconds and w the weight of dried sample in kg. The minimum detectable activity (MDA) of the present measurement system was calculated as follows (Currie 1968)

$$\mathrm{MDA} = \frac{\sigma\sqrt{\mathrm{B}}}{\varepsilon P t w}$$

where MDA is in Bq kg^{-1}, σ is the statistical coverage factor equal to 1.645 (confidence level 95%) and B is the background for the region of interest of a certain radionuclide. The MDA for ^{212}Pb and ^{40}K was 3.2 and 68.1 Bq kg^{-1}, respectively. Since ^{137}Cs peak was not observed in the background spectrum, the MDA for ^{137}Cs was not calculated.

Results and Discussion

Table 2 shows that the total radioactivity is formed mainly by isotopes of natural elements, chiefly ^{212}Pb, ^{40}K and ^{137}Cs, which were detected in the lichen specimens collected from Akçaabat (Trabzon) and Çamlıhemşin (Rize). The highest activities of the lichen species are seen in the locality of Helvacı (Trabzon, 100 m) and Çamlıhemşin (Rize, 1,850 m) while the activities are medium in Bozdoğan (Trabzon, 150 m), Kemaliye (Trabzon, 750 m) and Çamlihemşin (Rize, 900 m). Compared to Helvacı (Trabzon) and Çamlıhemşin (Rize, 1,850 m), medium activities of lichen species are seen in Bozdoğan (Trabzon), Kemaliye (Trabzon) and Çamlıhemşin (900 m). On the other hand, the activities of ^{137}Cs are very low in lichen species from Arpacılı–Akçaabat (Trabzon), Helvacı village-Akçaabat (Trabzon) and Bozdoğan village-Akçaabat-Trabzon (for *Cladonia furcata).* According to Table 2, the activity values generally increase significantly depending on the altitudes.

Soil permeability is important for the accumulation of radionuclides into species. It is well known that the most important parameters of the permeability are rainfall, the species and the pH values of the soil. In addition, as the surfaces of the species generally increase, they accumulate more radionuclides. The highest activities of ^{137}Cs were detected in foliose lichen species *Parmotrema perlatum* (Rize). The other foliose lichen species, *Flavoparmelia caperata* (Rize, Trabzon) and *Xanthoparmelia conspersa* (Trabzon), accumulate more radionuclides than the others except for *Diploschistes ocellatus.* The highest concentration of ^{137}Cs was detected in samples 4, 5, 6, 7, 11. In general, foliose and fruticose species, which have much more surfaces than those of crustose, absorb and accumulate more radionuclides than the others (Baysal and Yazıcı 1995, Papastefanou et al. 1989, Pipiska et al. 2007, Puhakainen et al. 2007, Ramzaev et al. 2007). The lichens, especially foliose lichens of *Parmelia*, *Flavoparmelia* and *Xanthoria*, can be used as bioindicator organisms to determine the radioactive pollution (Monnet et al. 2005, Weissman et al. 2006).

Different activity values were detected in *Cladonia furcata*, *C. portentosa*, *C. rangiformis, Flavoparmelia caperata* and *Xanthoparmelia conspersa*, although they were collected from the close localities. Thus, accumulation of the radionuclides can be attributed to the kind of species, their surfaces, anatomical properties, altitudes and wind. Additionally, various amounts of rain have fallen at different places after the Chernobyl accident in 1986.

Radioactive contaminants can spread not just across countries but also across continents as it was with the Chernobyl reactor accident. Therefore, radioactive levels in the air can change each month (Papastefanou et al. 2005). The lichen samples can accumulate the radioactive contaminants from air and substrata (soil, bark, rock etc.). Since the radioactive fallout of ^{137}Cs in the air can fall by rain and wind on different substrata, the place where the radioactive clouds and rainfall are high, more radioactive contaminants fall on lichen substrata. In this case, we noticed that the radioactive fallouts or contaminants in the air today are not very high as they were between 1986 and 1995 despite the high rainfall in Trabzon and Rize. Additionally, lichen species can reproduce permanently. It is known that young individuals can accumulate lower amounts of radioactive ^{137}Cs and ^{40}K. On the other hand, it must be taken into account that both different individuals (young) of the same species and radionuclides have half life. However, if the lichen specimens collected in 1995

Table 3 Radioactivity levels of lichens in 1995 (Baysal and Yazıcı 1995)

No	Samples	Locality	Bq kg^{-1} dry weight		
			^{134}Cs	^{137}Cs	^{40}K
1	*Cladonia furcata*	Akçaabat-Bozdoğan-TRABZON	14	685	1,936
2	*Cladonia portentosa*	Akçaabat-Bozdoğan-TRABZON	35	1,285	2,463
3	*Cladonia rangiformis*	Akçaabat-Bozdoğan-TRABZON	16	681	1,899
4	*Parmelia conspersa* (Syn. *Xanthoparmelia conspersa*)	Akçaabat-Bozdoğan-TRABZON	205	3,340	3,083
5	*Parmelia perlata* (Syn. *Parmotrema perlatum*)	Çamlıhemşin-RİZE	96	3,937	2,469
6	*Parmelia caperata*	Çamlıhemşin-RİZE	78	3,932	3,380
7	*Parmelia caperata* (Syn. *Flavoparmelia caperata)*	Akçaabat-Kemaliye-TRABZON	47	3,819	2,579
8	*Xanthoria parietina*	Akçaabat-Bozdoğan-TRABZON	80	1,027	4,066
9	*Xanthoria parietina*	Akçaabat-Helvacı-TRABZON	129	2,807	2,747
10	*Dermatocarpon miniatum*	Akçaabat-Helvacı-TRABZON	44	1,187	6,381
11	*Diploschistes ocellatus*	Akçaabat-Helvacı-TRABZON	27	2,445	3,407
12	*Peltigera praetextata*	Akçabat-Arpacılı-TRABZON	104	863	1,615
13	*Peltigera praetextata*	Çamlıhemşin-RİZE	46	530	1,519
14	*Lobaria pulmonaria*	Çamlıhemşin-RİZE	6	279	1,419

had been kept and radioisotopes of ^{134}Cs, ^{137}Cs and ^{40}K in them had been re-detected now, there might have been different radioactive levels of ^{134}Cs, ^{137}Cs and ^{40}K in proportion to present levels.

Potassium is one of the most important natural isotopes. The increase in ^{40}K radioactivity may come from two sources. The first one is connected with the natural presence of this isotope in soil, reaching 0.0119% of total potassium (UNSCEAR 1982). The highest potassium radioactivity was observed in lichen samples 5 and 9, located at Çamlıhemşin-Rize and Akçaabat-Trabzon but these values are lower than those of 1995 (Table 3). This is because of the anatomical features of *Parmotrema perlatum* and *Xanthoria parietina.* Additionally, it is thought that fine soil particles blown by winds are transported into the air and fall on the exposed parts of the plants and lichens and then were absorbed into their cells (Chibowski and Reszka 2001). Another increase of potasium concentration is connected with a long-term application of mineral fertilizing in these areas. Compared to 20 years ago, today there are not many agricultural and industrial sources hereabouts. But this kind of an agricultural area is seen in Arpacılı–Akçaabat-Trabzon, from which sample 3 and 12 were collected. Radioactive cesium isotopes ^{134}Cs and ^{137}Cs are very important to determine the level of the contamination of the environment. Both isotopes appear due to nuclear explosions and power plant failures. ^{134}Cs does not cause any problems and the concentration measured for this isotope is under the detection limit of the spectrometer equipped with Ge detector (Table 2).

Since the measurements were performed 20 years after the Chernobyl accident in 1986, radionuclides of ^{134}Cs having short half-life (2.062 y) have not been detected (L'Annunziata 2003). When the levels of the radionuclides in both studies are compared, the radioactivity levels of ^{137}Cs and ^{40}K in this study are significantly lower than those of 1995 (about 5.5–127 folds for ^{137}Cs, 5–17 folds for ^{40}K) (Fig. 2). Dangerous elements such as Cd and Cr were not detected and the level of ^{212}Pb was acceptably small. The atmospheric fallout of ^{137}Cs is very low in proportion to 1995 (Table 3). Therefore, the observed low activity may originate from a soil dust (Chibowski and Reszka 2001). Cesium absorbed by colloid soil particles is transported with them and may accumulate on the surfaces of the lichens. On the other hand, this isotope is kept in the heads of trees and then is washed down by rain. Therefore, lichens on barks may absorb this isotope on surfaces.

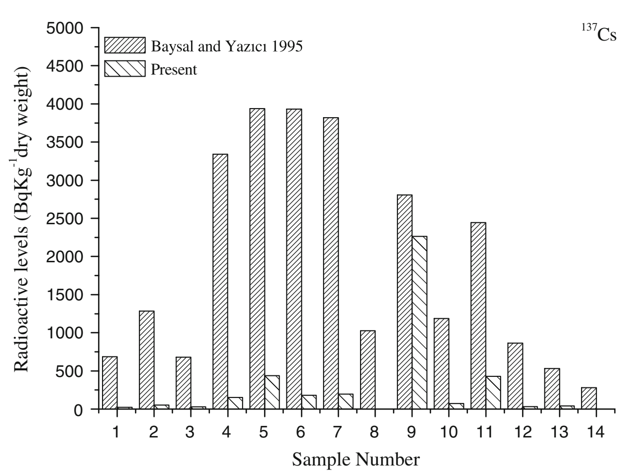

Fig. 2 Comparison of the present amounts of ^{137}Cs with those of 1995

References

Baysal A, Yazıcı K (1995) Radioactive contamination in lichens collected from Akçaabat and Çamlıhemşin in the eastern Black Sea region of Turkey, 1994. Toxicol Environ Chem 48:145–148

Currie LA (1968) Limits for qualitative detection and quantitative determination. Application to radiochemistry. Anal Chem 40:586–593. doi:10.1021/ac60259a007

Chibowski S, Reszka M (2001) Investigation of Lubnin town environment contamination byradionuclides and heavy metals with application of *Parmeliaceae* lichens. J Radioanal Nucl Ch 247:443–446. doi:10.1023/A:1006798828071

Çevik U, Damla N, Koz B, Kaya S (2008) Radiological characterization around Afsin Elbistan coal-fired power plant in Turkey. Energ Fuel 22:428–432

Gokmen IG, Akgoz M, Gokmen A (1996) Chernobyl radioactivity on the Black Sea coast of Turkey. Fresenius J Anal Chem 355:736–738

Golubev AV, Golubeva VN, Krylov NG, Kuznetsova VF, Mavrin SV, Aleinikov AY, Hoppes WG, Surano KA (2005) On monitoring anthropogenic airborne uranium concentrations and U-235/U-238 isotopic ratio by Lichen – bioindicator technique. J Environ Radioactiv 84:333–342. doi:10.1016/j.jenvrad.2005.04.002

Köse A, Topçuoğlu S, Varinlioğlu A, Kopya AI, Azar A, Uzun O, Karal H (1994) The levels of Caesium Radionuclides in Lichens in the Eastern Black Sea area of Turkey. Toxicol Environ Chem 45:221–224

Krstic D, Stevanovic N, Milivojevic J, Nikezic D (2007) Determination of the soil-to-grass transfer of Cs-137 and its relation to several soil properties at various locations in Serbia. Isot Environ Healt S 43:65–73. doi:10.1080/10256010601154171

LaBrecque JJ, Cordoves PR (2007) Determination and spatial distribution of ^{137}Cs in soils, mosses and lichens near Kavanayen, Venezuela. J Radioanal Nucl Ch 273:401–404. doi:10.1007/s10967–007-6847-2

L'Annunziata MF (2003) Handbook of radioactivity analysis. 2nd edn. Academic Press, London

McClenahen JR, Davis DD, Hutnik RJ (2007) Macrolichens as biomonitors of air-quality change in western Pennsylvania. Northeast Nat 14:15–26. doi:10.1656/1092-6194(2007)14[15:MABOAC]2.0.CO;2

Monnet F, Bordas F, Deluchat V, Chatenet P, Botineau M, Baudu M (2005) Use of the aquatic lichen *Dermatocarpon luridum* as bioindicator of copper pollution: accumulation and cellular distribution tests. Environ Pollut 138: 455–461. doi:10.1016/j.envpol.2005.04.019

Paatero J, Kulmala S, Jaakkola T, Saxen R, Buyukay M (2007) Deposition of Sb-125, Ru-106, Ce-144, Cs-134 and Cs-137 in Finland after the Chernobyl accident. Boreal Env Res 12:43–54

Papastefanou C, Manolopoulou M, Sawidis T (1989) Lichens and Mosses Biological monitors of radioactivity fallout the Chernobyl reactor accident. J Environ Radioactiv 9:199–207. doi:10.1016/0265-931X(89)90044-1

Papastefanou C, Manolopoulou M, Stoulos S, Ioannidou A, Gerasopoulos E (2005) Cesium-137 in air late after the Chernobyl reactor accident. J Radional Nucl Chem Lett 264:699–700. doi:10.1007/s10967-005-0773-y

Pipiska M, Hornik M, Vrtoch L, Augustin J, Lesny J (2007) Biosorption of Co2+ ions by lichen *Hypogymnia physodes* from aqueous solutions. Biologia 62:276–282. doi:10.2478/s11756-007-0047-y

Puhakainen M, Rahola T, Heikkinen T, Illukka E (2007) Cs-134 and (CS)-C-137 in lichen (*Cladonia stellaris*) in southern Finland. Boreal Env Res 12:29–35

Ramzaev V, Mishine A, Kaduka M, Basalaeva L, Brown J, Andersson KG (2007) Cs-137 and Sr-90 in live and dead reindeer lichens (genera *Cladonia*) from the "Kraton-3" underground nuclear explosion site. J Environ Radioactiv 93:84–99. doi:10.1016/j.jenvrad.2006.11.008

Rossbach M, Lambrecht S (2006) Lichens as biomonitors: global, regional and local aspects. Croat Chem Acta 79:119–124

Saka AZ, Çevik U, Bacaksız E, Kopya AI, Tıraşoğlu E (1997) Levels of caesium radionuclides in lichens and mosses from the province of Ordu in the Eastern Black Sea area of Turkey. J Radional Nucl Chem Lett 222:87–92. doi:10.1007/BF02034252

Skuterud L, Gaare E, Eikelmann IM, Hove K, Steinnes E (2005) Chernobyl radioactivity persists in reindeer. J Environ Radioactiv 83:231–252. doi:10.1016/j.jenvrad.2005.04.008

Topcuoglu S, Gungor N, Köse A, Varinlioglu A (1997) Translocation and depuration of ^{137}Cs in tea plants. J Radioanal Nucl Ch 218:263–266. doi:10.1007/BF02039348

United Nations Scientific Committee on the Effects of Atomic Radiation (1982) On the effects of atomic radiation: ionizing radiation sources and biological effects, New York

Weissman L, Fraiberg M, Shine L, Garty J, Hochman A (2006) Responses of antioxidants in the lichen *Ramalina lacera* may serve as an early-warning bioindicator system for the detection of air pollution stres. Fems Microbiol Ecol 58:41–53. doi:10.1111/j.1574-6941.2006.00138.x