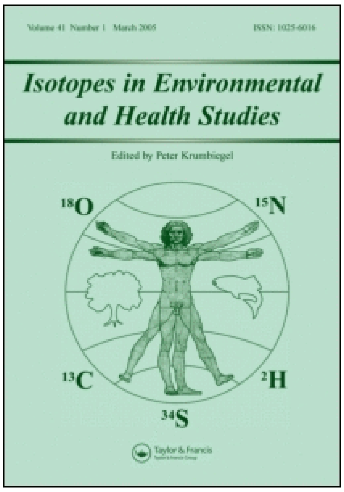

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# $^{137}$Cs and $^{40}$K activity concentration measurements and elemental analysis in lichen samples collected from the Giresun province of northeastern Turkey

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# $^{137}$Cs and $^{40}$K activity concentration measurements and elemental analysis in lichen samples collected from the Giresun province of northeastern Turkey

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About 21 years after the Chernobyl accident, $^{137}$Cs and $^{40}$K activity concentration measurements using gamma-ray spectroscopy and elemental analysis using energy dispersive X-ray spectroscopy were performed in five different lichen species collected from the Giresun province of northeastern Turkey. Being a symbiosis of algae and fungi, lichens are mostly used for environmental measurements since the fungal partner is responsible for the uptake of necessary nutrients or harmful substances, such as heavy metals of radionuclides. The gamma activity results showed that $^{137}$Cs, an artificial radionuclide released from the Chernobyl power plant accident, is still eminent in the environment of the province. The mean activity concentrations of $^{137}$Cs and $^{40}$K ranged from 24 to 254 with the mean value of 102 Bq kg$^{-1}$ and from 345 to 2103 with the mean value of 1143 Bq kg$^{-1}$ in dry weight. The results of the elemental analyses showed potassium, calcium, titanium, iron, tin, and barium in different concentrations.

**Keywords:** cesium-137; elemental analysis; lichens; natural radioactivity; potassium-40; Turkey

## 1. Introduction

As a result of nuclear explosions carried out in the earth's atmosphere and the Chernobyl nuclear power station accident, the world has become polluted with radionuclides of artificial origin. The radionuclides that caused substantial radiopollution were $^{137}$Cs, $^{134}$Cs, and $^{131}$I. Due to the short physical half-life of $^{131}$I (8 d), the environmental pollution by this radionuclide was of a temporary nature, without diminishing the importance of the damage caused to biological systems. To some extent, the same is true for $^{134}$Cs, which has a half-life of 2 y. On the other hand, $^{137}$Cs with a half-life of 30.18 y has remained in the ecosystems many years after the accident.

The Chernobyl disaster had an impact on many countries. Regions affected by radioactive fallout included not only Ukraine itself but also Belarus, Georgia, Poland, Sweden, Germany, Austria, Hungary, Finland, Norway, Turkey, and others. After this accident, intensive large-scaled radiological investigations of the environmental samples have been started worldwide. The radioactive plume from the Chernobyl accident of 26 April 1986 reached Turkey by 5 May, substantially contaminating various regions and ecosystems of the country. The northeastern part of Turkey was

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one of the most seriously contaminated regions by this tragic accident [1]. During the emergency, Cekmece Nuclear Research and Training Center performed an analysis of various substances. In their report, it has been noted by Gedikoglu and Sipahi [2] that the level of $^{137}$Cs activity concentration in the Turkish tea plant was found to be a maximum value of 44,000 Bq kg$^{-1}$ for the 1986 products, and 373 Bq kg$^{-1}$ activity level was reported elsewhere for $^{137}$Cs in lichen samples collected from the Giresun province in 1997 [3].

$^{40}$K is a natural radionuclide with a half-life of $1.3 \times 10^9$ y, which is part of environmental background radiation. Isotopes of caesium are chemical analogues of potassium and, consequently, they are included in the nutrient cycle and cause long-term irradiation to biota and man [4].

Atmospheric particulates have attracted a great environmental concern over the last few decades because of the evidence that this type of pollution can have severe long-term implications for respiratory illness in humans [5]. In particular, heavy metals adsorbed on ambient particles were found to cause damage to lung tissues [6]. Due to rapid urbanisation and industrial development in recent years, atmospheric pollution has caused serious deterioration of the terrestrial environment in many countries. Thus, the monitoring of airborne metals in the urban environment has become an essential part of environmental planning and control programmes in many parts of the world.

Lichens being the effect of fungus and algae symbiosis, where both organisms construct the thallus, a new organism of a specific structure and physiology, are a very useful group of organisms in the environmental monitoring. Because of the lack of any organs to assimilate water and mineral salts from the subsoil, these organisms assimilate all their nourishment from the air and thus may be excellently utilised for investigations of the atmosphere contamination, including the radioactive fall-out determination. The elements and substances assimilated from the air by the thallus of the lichens are accumulated in them, particularly without the possibility of desorption [7]. More pertinently, lichens were used as biomonitors by many researchers in the wake of the Chernobyl accident [8–13].

As shown in Figure 1, the Giresun province is located in the northeastern part of Turkey between the longitudes of 37°50′–39°12′E and the latitudes of 40°07′–41°08′N, with a total population of 524,000. The weather is semi-cold sub-humid with temperature variations between 7 and 22 °C. The average annual temperature is 11 °C, and the average annual precipitation is 140 mm [14]. Although the province is a very attractive place, unfortunately there are hardly any comprehensive studies or scientific data at any official or academic institutions in Turkey showing the environmental situation in this province.

The aim of the present study is to define the contamination level of the studied area by measuring $^{137}$Cs activity concentrations 21 y after the Chernobyl accident using gamma-ray spectroscopy and to make an elemental analysis using energy dispersive X-ray spectroscopy (EDXRF) in lichen samples collected for the studied area. $^{137}$Cs activity concentration data obtained are expected to serve as baseline data to estimate an important parameter of ecological half-life of $^{137}$Cs in this region for future studies.

## 2. Experimental method

### 2.1. $^{137}$*Cs and* $^{40}$*K activity measurements*

Twenty one years after the Chernobyl accident, the lichen samples were collected from the studied area and dried in an oven at a temperature of 70 °C, sieved to remove stones and pebbles, and crushed in the laboratory to homogenise them. Then each sample was measured, and the results are given in Bq kg$^{-1}$ dry weight.

The gamma-ray activities were measured using high-purity germanium computer controlled detector having a resolution of 2 keV for the 1332 keV energy line of $^{60}$Co with conventional electronics and 20% relative efficiency, and Genie 2000 as software. The detector was shielded

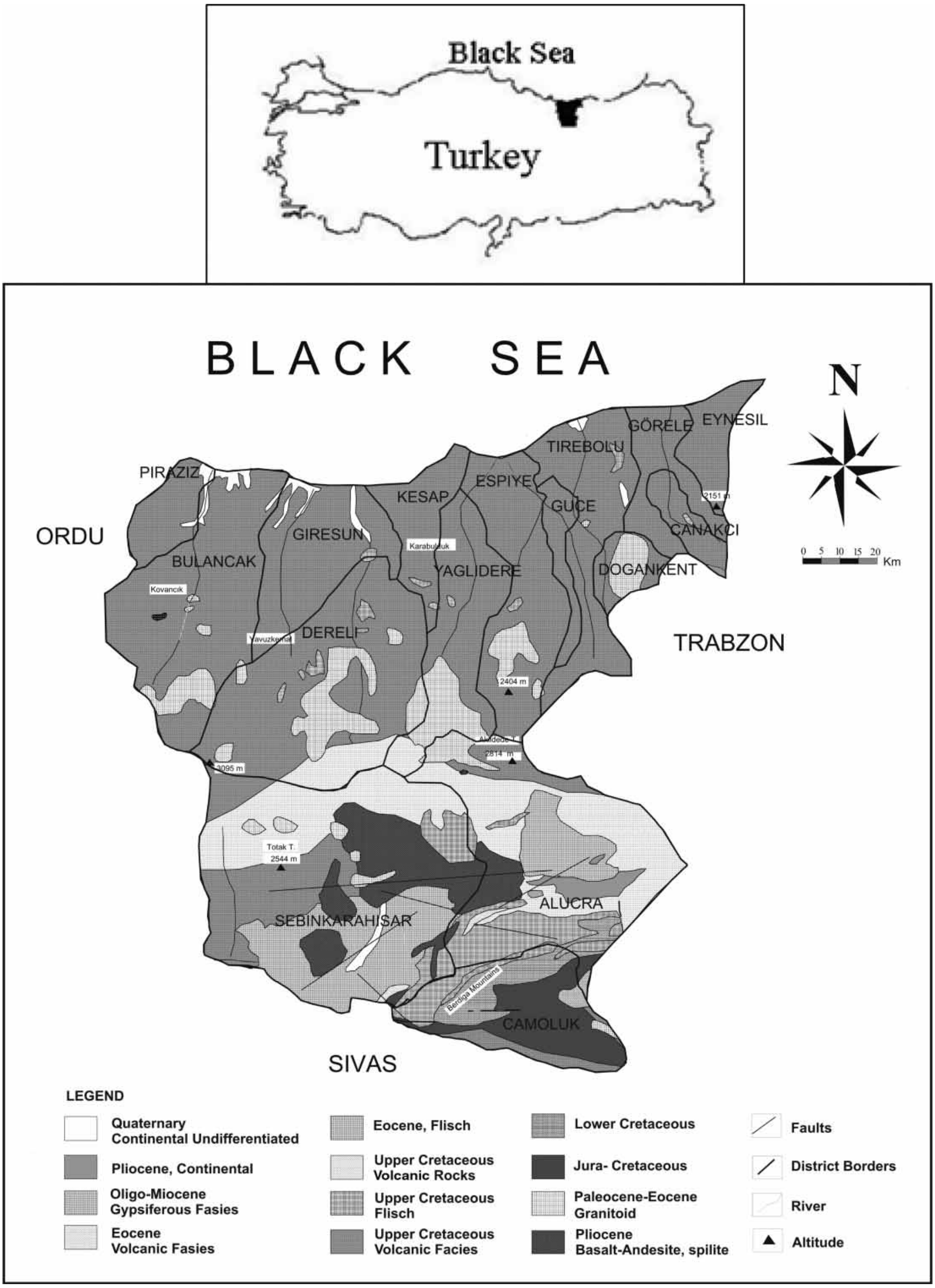

Figure 1. The geological map of the studied area.

with a 10 cm thick lead layer to reduce the background due to the cosmic rays and the radiation nearby the system.

Full-energy peak efficiency was determined using the Standard Reference Material prepared by the International Atomic Energy Agency [15]. Decay corrections were performed to the sampling date. The activity of $^{137}$Cs and $^{40}$K was determined using their 661 and 1460 keV gamma-ray lines, respectively.

### 2.2. *Elemental analysis using EDXRF*

Quantitative analysis for the elements was carried out using the standard addition method using EDXRF. The method involves the addition of known quantities of analyte to the specimen. If the analyte is presented at low levels and no suitable standards are available, standard addition may prove to be an alternative, especially if the analyst is interested in only one analyte element. Certain amounts of the element to be analyzed are added to samples.

All samples were dried in a Heraeus furnace and then ground in a spex mill. To reduce particle size effect, the powder obtained was sieved using a 400 mesh sieve and then stirred for 25 min to obtain a well mixed sample. Forty milligrams of the powder prepared was pressed in 13 mm diameter.

Samples were irradiated by 59.5 keV photons emitted by an annular 50 mCi $^{241}$Am radioactive source for iron, tin, and barium determination and irradiated by 5.9 keV photons emitted by an annular 50 mCi $^{55}$Fe radioactive source for potassium, calcium, and titanium. A Princeton Gamma-Tech Si(Li) detector having 147 eV full-width at half-maximum for 5.9 keV was used for the experiment. For data acquisition, 2048 channels of a multi channel analyzer were employed. In qualitative analysis, characteristic X-rays emitted by excited atoms of the sample were registered for 2000 s. The K content was also estimated on the assumption that the natural $^{40}$K abundance (0.0117%) is equivalent to a relation $^{40}$K(Bq)/K(g) = 30.3 Bq g$^{-1}$ [16,17].

In elemental analysis using X-ray fluorescence technique, matrix effects are known to distort the linearity of ‘photo peak area versus concentration’ graphs for analyses [18]. To minimise or to eliminate matrix effects, the $K_\alpha$ net peak areas for iron, tin, and barium obtained from sample spectra were normalised by dividing them by Compton net peak areas. The $K_\alpha$ net peak areas for potassium, calcium, and titanium obtained from sample spectra were normalised by dividing them by Mn$K_\alpha$ peak areas.

## 3. Results and discussion

### 3.1. *$^{137}$Cs and $^{40}$K concentration results*

$^{137}$Cs and $^{40}$K activity concentration results in lichen samples collected from the studied area are given in Table 1. As can be seen from the table, $^{137}$Cs activity concentrations ranged from 24 to

Table 1. $^{137}$Cs and $^{40}$K activity concentrations of the lichen samples.

| Sampling area | Species of the lichens | $^{137}$Cs (Bq kg$^{-1}$) | $^{40}$K (Bq kg$^{-1}$) | $^{137}$Cs/$^{40}$K |
|---|---|---|---|---|
| Giresun (Centre) | *Flavoparmelia caperata* | 210 ± 41 | 1324 ± 123 | 0.16 |
| Piraziz | *Xanthoparmelia conspersa* | 76 ± 18 | 1205 ± 86 | 0.06 |
| Bulancak | *Flavoparmelia caperata* | 120 ± 43 | 1336 ± 137 | 0.09 |
| Keşap | *Flavoparmelia caperata* | 127 ± 45 | 768 ± 101 | 0.17 |
| Yaglidere | *Physcia adscendes* | 175 ± 13 | 656 ± 35 | 0.27 |
| Espiye | *Physcia adscendes* | 24 ± 05 | 1291 ± 153 | 0.02 |
| Guce | *Parmotrema perlatum* | 254 ± 18 | 1302 ± 94 | 0.20 |
| Tirebolu | *Flavoparmelia caperata* | 27 ± 10 | 1315 ± 122 | 0.02 |
| Gorele | *Flavoparmelia caperata* | 66 ± 09 | 1381 ± 159 | 0.05 |
| Canakci | *Xanthoria parietina* | 167 ± 32 | 709 ± 123 | 0.24 |
| Eynesil | *Physcia adscendes* | 150 ± 54 | 1400 ± 154 | 0.11 |
| Dogankent | *Parmotrema perlatum* | 73 ± 17 | 970 ± 231 | 0.08 |
| Dereli | *Flavoparmelia caperata* | 176 ± 35 | 2103 ± 242 | 0.08 |
| Sebinkarahisar | *Flavoparmelia caperata* | 80 ± 19 | 1320 ± 129 | 0.06 |
| Alucra | *Physcia adscendes* | 120 ± 21 | 345 ± 59 | 0.35 |
| Camoluk | *Flavoparmelia caperata* | 93 ± 22 | 867 ± 121 | 0.11 |
| Mean | – | 102 ± 30 | 1143 ± 140 | 0.09 |

254 with the mean value of 102 Bq kg$^{-1}$, and $^{40}$K activity concentrations ranged from 345 to 2103 with the mean value of 1143 Bq kg$^{-1}$ in dry weight. The Espiye district showed the lowest value and the Guce district showed the highest value in $^{137}$Cs activity concentration. On the other hand, the Alucra district showed the lowest value and the Dereli district showed the highest value in $^{40}$K activity concentration. In all the species studied in the present work, the specific activity of $^{40}$K was higher than that of $^{137}$Cs. Since potassium has practically the same physicochemical properties as cesium, some authors suggested that $^{137}$Cs might be taken up by the lichens as a surrogate for

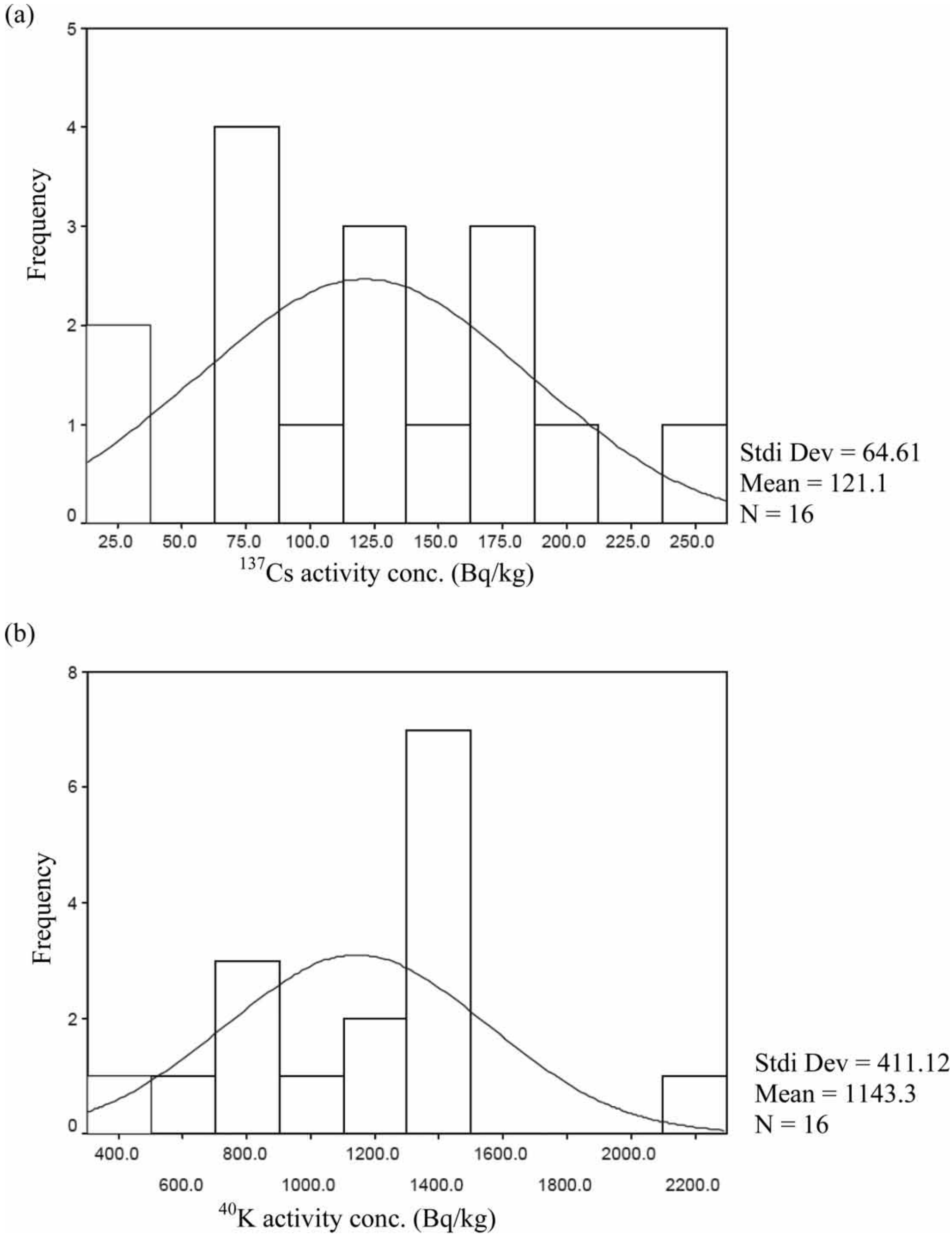

Figure 2. Frequency distributions of (a) $^{137}$Cs and (b) $^{40}$K activity concentrations.

potassium [9,11]. However, the statistical analysis (Pearson correlation) showed that there is no correlation between $^{137}$Cs and $^{40}$K activity concentrations in the lichen samples in the present study. On the other hand, excluding the extremely high values of $^{40}$K activity concentrations, one can see that the activity concentrations of $^{40}$K decrease with increasing $^{137}$Cs activity.

An SPSS computer software was used to compute ordinary statistics. For the collected samples, the frequency distributions of $^{137}$Cs and $^{40}$K activities are represented in Figure 2 and their fits are plotted considering the skewness and the kurtosis coefficients which show the distribution of variables whether it obeys the normal or the lognormal law. It was found that $^{137}$Cs and $^{40}$K activity concentration distributions in lichen samples follow an approximately normal pattern (Figure 2). The skewness and the kurtosis coefficients for frequency distributions of $^{137}$Cs and $^{40}$K are 0.361, $-0.355$ and 0.176, 1.125, respectively. The positive values of skewness coefficients indicate that the distributions are asymmetric, with the right tail longer than the left. On the other hand, the negative value of the kurtosis indicates just the opposite. The low kurtosis coefficients suggest that the distribution is close the normal.

The results show that $^{137}$Cs radionuclide is still eminent in the environment of this province. Figure 3 shows the interpolated $^{137}$Cs activity concentration map of the province. The complex dispersion pattern of the activity concentration could be attributed to the changing meteorological conditions during the active phase of the accident with winds from different directions [19].

### 3.2. *Elemental analysis results*

Qualitative analysis of spectral peaks showed that the samples contained potassium, calcium, titanium, iron, tin, and barium (Table 2). The mean concentrations of K, Ca, Ti, Fe, Sn, and Ba in different lichen samples were measured to be 8.57, 24.12, 7.24, 25.39, 0.14, and 0.39 mg g$^{-1}$, respectively. The higher or the lower concentration of elements detected could be related to morphological and anatomical features of lichen, such as metal-absorbing abilities, surface area,

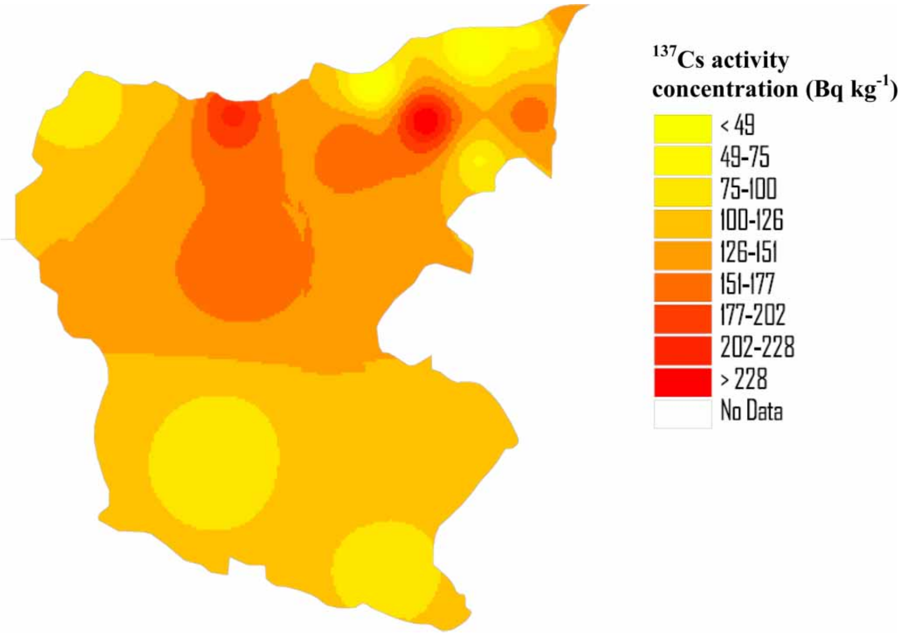

Figure 3. The interpolated map of $^{137}$Cs activity concentration in lichen samples collected from the studied area.

Table 2. Elemental concentrations in lichen samples (mg $g^{-1}$).

| Sampling area | Species of lichen | K[†] | K[‡] | Ca | Ti | Fe | Sn | Ba |
|---|---|---|---|---|---|---|---|---|
| Giresun(centre) | *Flavoparmelia caperata* | 3.46 | 43.70 | 37.48 | 5.74 | 18.61 | 0.06 | 0.28 |
| Piraziz | *Xanthoparmelia conspersa* | 11.15 | 39.77 | 26.72 | 6.48 | 16.59 | 0.09 | 0.39 |
| Bulancak | *Flavoparmelia caperata* | 6.01 | 44.09 | 20.13 | 5.37 | 93.85 | 0.03 | 0.3 |
| Keşap | *Flavoparmelia caperata* | 13.02 | 25.35 | 8.69 | 6.1 | 15.6 | 0.24 | 0.17 |
| Yaglidere | *Physcia adscendes* | 3.46 | 21.65 | 23.21 | 5.9 | 19.53 | 0.16 | 0.22 |
| Espiye | *Physcia adscendes* | 12.92 | 42.61 | 9.02 | 9.44 | 17.62 | 0.04 | 0.63 |
| Guce | *Parmotrema perlatum* | 9.04 | 42.97 | 5.66 | 5.95 | 34.01 | 0.32 | 0.14 |
| Tirebolu | *Flavoparmelia caperata* | 2.03 | 43.40 | 51.6 | 3.9 | 36.36 | 0.06 | 0.14 |
| Gorele | *Flavoparmelia caperata* | 5.31 | 45.58 | 30.21 | 8.28 | 19.77 | 0.21 | 0.5 |
| Canakci | *Xanthoria parietina* | 11.18 | 23.40 | 9.91 | 12.44 | 13.21 | 0.11 | 0.68 |
| Eynesil | *Physcia adscendes* | 12.36 | 46.20 | 11.25 | 8.69 | 15.32 | 0.09 | 0.47 |
| Dogankent | *Parmotrema perlatum* | 7.33 | 32.01 | 21.66 | 6.89 | 22.95 | 0.17 | 0.29 |
| Dereli | *Flavoparmelia caperata* | 12.18 | 69.41 | 29.24 | 5.74 | 13.72 | 0.14 | 0.58 |
| Sebinkarahisar | *Flavoparmelia caperata* | 3.07 | 43.56 | 74.05 | 6.31 | 27.45 | 0.15 | 0.34 |
| Alucra | *Physcia adscendes* | 14.55 | 11.39 | 13.05 | 10.49 | 10.13 | 0.29 | 0.72 |
| Camoluk | *Flavoparmelia caperata* | 10.03 | 28.61 | 14.01 | 8.11 | 16.12 | 0.12 | 0.4 |
| Mean | – | 8.57 | 37.73 | 24.12 | 7.24 | 25.39 | 0.14 | 0.39 |

[†] Measured values.
[‡] Estimated values.

large intercellular space, and high cell membrane permeability. Also these variations could be related with environmental factors as pH, additives, elemental concentrations in the atmosphere, humidity, direction of dominating wind, and other climatic conditions that can have various effects on metal concentration in lichens.

The predicted and measured K concentrations are given in Table 2. All the estimated values (except for K concentration in *Physica adscendes*) are higher than the measured values. Big differences between the estimated and measured K element concentration values indicate that the present data do not confirm the assumption that the $^{40}$K/K ratio is constant in nature.

Ba and Ca were classified as important components in brakes by some authors [20,21]; hence, the existence of these metals could be attributed to the traffic in the province. Ca is predominantly an indicator for soil dust, as Ca is a frequent component of grit used on many roads during winter. Fe and Sn are heavy metals. It has been observed that the mean concentration of Fe is higher than that of Sn (Table 2). Pb was not detected in any lichen species in the present study. This could be because of the detection limit of our system: the detection limit was defined to be 5 mg $kg^{-1}$ for Pb elsewhere [22]. The Pearson correlation was performed to obtain potential elemental correlations from the data set (Table 3). As a result, strong correlations were observed between (Ca–K), (Ti–K), (Ba–K), and (Ba–Ti), indicating their potential anthropogenic inputs.

Table 3. The Pearson correlation coefficient matrix between element concentrations in the lichens samples.

| | K | Ca | Ti | Fe | Sn | Ba |
|---|---|---|---|---|---|---|
| K | 1.000 | | | | | |
| Ca | 0.002** | 1.000 | | | | |
| Ti | 0.018* | 0.064 | 1.000 | | | |
| Fe | 0.136 | 0.677 | 0.095 | 1.000 | | |
| Sn | 0.350 | 0.351 | 0.666 | 0.253 | 1.000 | |
| Ba | 0.016* | 0.348 | 0.000** | 0.153 | 0.824 | 1.000 |

**$P < 0.01$.
*$P < 0.05$.

In our measurements, maximum relative errors due to the counting system were of the order ~0.5–5%. Errors originating from sample weighing, source intensity, and system geometry were about 4%. The combined relative error in our results was accordingly of the order of 8%.

## 4. Conclusion

$^{137}$Cs and $^{40}$K activity concentration measurements and elemental analysis have been carried out in some lichen species collected from the Giresun province of northeastern Turkey. It was found that $^{137}$Cs is still eminent in the environment of the province. An interpolated map of the studied area was created in order to visualise the contamination level and contaminated regions due to the $^{137}$Cs release from the power plant.

Due to the detection limit of our system (EDXRF), Pb was observed in none of the lichen species collected. Another experimental set-up having a suitable detection limit might be used to measure Pb.

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