



## Effect of air pollution on element profile and radioactive compounds in six tree species

### Altı ağaç türünde hava kirliliğinin element profili ve radyoaktif bileşikler üzerine etkisi

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#### Abstract

Variation in macro- and micro-nutrients and radioactivities (<sup>238</sup>U, <sup>232</sup>Th, <sup>137</sup>Cs, and <sup>40</sup>K) in fresh leaf samples due to air pollution was studied using six tree species: maple (*Acer negundo*), cypress (*Cupressus arizonica*), ash-tree (*Fraxinus excelsior*), pine (*Pinus nigra*), plane tree (*Platanus orientalis*), and poplar (*Populus nigra*). The leaf samples were collected in 2016 and 2017 from a rural area (control site) and urban area (polluted site) which had high population, heavy traffic, and small industrial activity. The results showed that both 2016 and 2017, mean Mg, S and K concentrations in the leaf samples were higher at the polluted sites than at the control sites for all tree species, except for ash tree for Mg, ash and plane trees for S and maple tree for K. Mean P concentration was lower at the polluted sites for maple, pine, plane and poplar trees, while it was higher for cypress and ash trees. Mean Ca concentration at the polluted sites was lower for maple, cypress, pine and plane trees, but higher for ash and poplar trees. Most micronutrients were generally higher at the polluted sites than at the control sites for all tree species. Only, Co concentration at the polluted site was lower for ash and pine trees than at the control site, while Mn concentration was lower for poplar tree at the polluted sites. The activity concentration of trees was recorded an increasing order as <sup>40</sup>K > <sup>232</sup>Th > <sup>238</sup>U > <sup>137</sup>Cs in 2016, while they were noted as <sup>40</sup>K > <sup>238</sup>U > <sup>232</sup>Th > <sup>137</sup>Cs in 2017. The plane tree had the lowest activity concentration for both years. Taking all data into an account, the order of trees at the polluted sites was determined as poplar > ash tree > maple > pine > plane tree = cypress for 2016, while this order was recorded as poplar > cypress = maple > ash tree > pine = plane tree for 2017. It can be concluded that poplar, ash tree, and maple could be recommended for future biomonitoring research in order to improve air quality in urban areas.

**Keywords:** Air pollution, nutrients, radionuclide, trees, Kastamonu

#### Özet

Yeşil yaprak örneklerinin makro ve mikro besin maddeleri ile radyoaktivitelerinin (<sup>238</sup>U, <sup>232</sup>Th, <sup>137</sup>Cs ve <sup>40</sup>K) hava kirliliğine bağlı değişimi altı farklı ağaç türünde incelenmiştir: akçaağaç (*Acer negundo*), selvi (*Cupressus arizonica*), dişbudak (*Fraxinus excelsior*), karaçam (*Pinus nigra*), çınar (*Platanus orientalis*), and kara kavak (*Populus nigra*). Yaprak örnekleri, 2016 ve 2017 yıllarında, yüksek nüfus, yoğun trafik ve küçük endüstriyel aktiviteye sahip bir kırsal alandan (kontrol alanı) ve kentsel alandan (kirlenmiş alan) toplanmıştır. Sonuçlar, hem 2016 hem de 2017'de, yaprak örneklerindeki ortalama Mg, S ve K konsantrasyonlarının, Mg için dişbudak, S için dişbudak ve çınar, K için akçaağaç türleri hariç, diğer tüm ağaç türleri için kirlenmiş bölgelerde kontrol bölgelerine göre daha yüksek olduğunu göstermiştir. Kirlenmiş alanlarda ortalama P konsantrasyonu akçaağaç, çam, çınar ve kavak ağaçlarında daha düşük, selvi ve dişbudak ağaçlarında daha yüksek bulunmuştur. Kirlenmiş alanlarda ortalama Ca konsantrasyonu akçaağaç, selvi, çam ve çınar ağaçları için daha düşük, dişbudak ve kavak ağaçları için daha yüksek çıkmıştır. Mikrobisim elementlerinin çoğu, tüm ağaç türleri için genellikle kirlenmiş bölgelerde kontrol bölgelerine göre daha yüksek belirlenmiştir. Sadece, kirlenmiş sahadaki Co konsantrasyonu, kontrol sahasına göre dişbudak ve çam ağaçları için daha düşük belirlenirken, kirlenen sahalardaki kavak ağacı için Mn konsantrasyonu daha düşük bulunmuştur. Ağaçların aktivite konsantrasyonu 2016 yılında artan bir sırayla <sup>40</sup>K > <sup>232</sup>Th > <sup>238</sup>U > <sup>137</sup>Cs olarak belirlenirken, 2017 yılında <sup>40</sup>K > <sup>238</sup>U > <sup>232</sup>Th > <sup>137</sup>Cs olarak belirlenmiştir. Her iki yılda da en düşük aktivite konsantrasyonuna çınar ağacı sahip olmuştur. Tüm veriler dikkate alındığında, 2016 yılı için kirlenmiş alanlardaki ağaçların sıralaması kavak > dişbudak > akçaağaç > çam > çınar = selvi olarak belirlenirken, bu sıralama 2017 için kavak > selvi = akçaağaç > dişbudak > çam = çınar ağacı olarak sıralanmıştır. Sonuç olarak, kentsel alanlarda hava kalitesini iyileştirmek için gelecekteki biyozileme araştırmaları için kavak, dişbudak ve akçaağaç türleri önerilebilir.

**Anahtar kelimeler:** Hava kirliliği, besin elementleri, radyoaktivite, ağaç, Kastamonu

## 1. Introduction

Nowadays, the increasing urbanization that has occurred throughout the world, is one of the issues that contribute to the development of environmental problems. The rapid expansion of cities due to the shift from rural to urban results in an increase in population density, house and vehicles number, industrial activities, consumption of fossil-fuel burning, shrinking green areas due to the land expansion, and overall human adverse impacts (Baycu et al., 2015). All the consequences of urbanization have led to an increase in local air pollution together with climate change and various diseases (Turfan et al., 2018; Arıcak et al., 2020). It has been reported that over one half of the world's population are now living in urban areas and 60-90% of the world's population will inhabit urban areas by 2030 (HEI, 2018). This means that the detrimental effects of air pollution on the ecosystem will continue to increase even more in urban. It is known that pollutants contain various gasses (SO<sub>2</sub>, CO/CO<sub>2</sub>, NO), particulates, heavy metals, aerosols, and also natural and artificial radioactive elements (40K, <sup>226</sup>Ra, <sup>238</sup>U, <sup>232</sup>Th, and <sup>137</sup>Cs) which can result in catastrophic effects on the sustainability and biodiversity of urban plants (Wei et al., 2017; Yümün et al., 2021). However, urban forests are dynamic urban ecosystems that provide critical benefits to people and wildlife. They contribute to maximizing local air quality by altering the deposition, dispersion rates of pollutants by leaves and barks, especially where air pollution can pose public health risks (Baycu et al., 2015). Therefore, they can be used to evaluate whether certain ecophysiological responses may be useful biomarkers of urban pollution. However, several factors need to be taken into account in the selection of plant species for remediation and improving air quality. For example, larger trees with large leaf surfaces are excellent filters for urban air pollutants. It has been reported that evergreen trees, in particular, coniferous as *Pinus*, *Cupressus*, and *Taxus* and broadleaf taxa like *Acer*, *Aesculus*, *Fraxinus*, *Platanus*, *Populus*, and *Robinia* are good sources for remediation observations (Aksoy and Demirezen, 2006; Petrova et al., 2014). There are a number of extensive studies

related to the identification of resistant species to pollution as bioindicator species in Kastamonu province (Arıcak et al., 2020; Cetin et al., 2020; Saleh, 2018). However, there are a few studies investigated the effect of air pollution on the accumulation of nutrients, heavy metals, and radionuclides in tree leaves. The purpose of this study was to reveal the nutrient status and the activity concentrations of 40K, <sup>226</sup>Ra, <sup>238</sup>U, <sup>232</sup>Th, and <sup>137</sup>Cs in the leaves of maple (*Acer negundo*), cypress (*Cupressus arizonica*) ash tree (*Fraxinus excelsior*), pine (*Pinus nigra*), plane tree (*Platanus orientalis*), and poplar (*Populus nigra*), which are the most abundant taxa grown in parks and roadsides in the city centre of Kastamonu, Turkey.

## 2. Materials and Methods

### 2.1. Description of study area

This study was conducted in Kastamonu, located northwest of Turkey (41°22' N-33°47' E) (Figure 1). The mean altitude of Kastamonu city center is 791 m. Kastamonu is located within a transition zone that experiences both oceanic and continental climates. Kastamonu has a small stream called Karaçomak, which runs through the city center. The main transportation of the city runs on both east and west side roads of this stream (Figure 1). Kastamonu is one of the most important provinces in the Black Sea Region with its natural beauty and cultural richness. However, Kastamonu is among the second degree polluted cities due to the migrations (İbret, 2011; Cetin et al., 2020). Those researchers have stated that the rural-urban migration has increased the number of vehicles, houses in the city centres as well as natural gas consumption in Kastamonu city center. Study sites were chosen at two areas as rural area (control) and urban area (polluted) shown in Figure 1. Two control sites were in the garden of the 15<sup>th</sup> Regional Directorate of Highways, which was far from the polluted area (Figure 1), while the polluted sites were at both sides of the road D765 at the beginning and the end of the city with heavy traffic due to the most of the official institutions, high population density and being the first central settlement (Duman, 2017; Gemici, 2017).

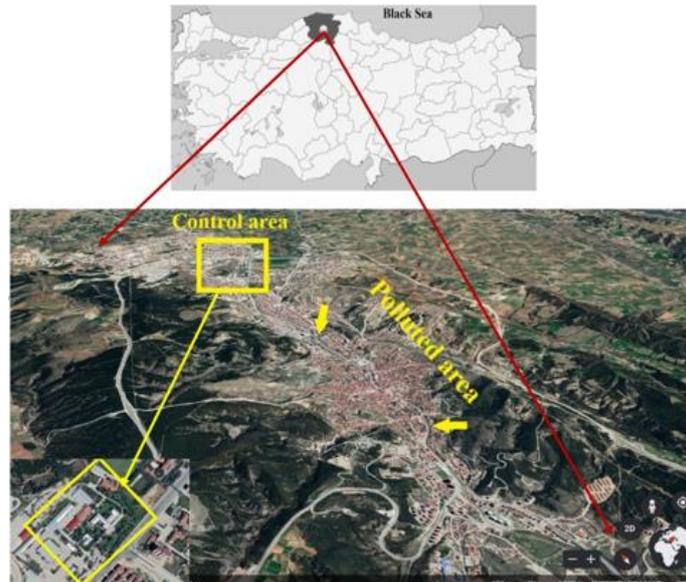


Figure 1. Location of Kastamonu and the sampling sites as polluted and unpolluted (control) areas within the city border of Kastamonu

Table 1. Some silvicultural characteristics of tree species

Name of trees	Age (Year)	Height (m)	Diameter (cm)	Number of trees
Maple ( <i>Acer negundo</i> L.)	15-20	8-10	25-30	8
Cypress ( <i>Cupressus arizonica</i> L.)	10-15	10-12	20-22	10
Ash ( <i>Fraxinus excelsior</i> L.)	15-20	10-12	25-30	8
Pine ( <i>Pinus nigra</i> L.)	15-20	8-10	22-25	10
Plane ( <i>Platanus orientalis</i> L.)	15-20	10-12	25-30	10
Poplar ( <i>Populus nigra</i> L.)	10-15	13-15	20-25	8

Pollution level of Kastamonu is observed from the Air Quality Assessment and Management Station (AQAMS). According to data, in the dispersed SO<sub>2</sub>, 76% was scattered as 0-5 µg/m<sup>3</sup>, 22.8% as 5-10, and 4-5% as 10-55 µg/m<sup>3</sup>. 7.5 % of the dust concentrations (PM10) ranged from 1 to 10 µg/m<sup>3</sup>, 26.6% from 10 and 20 µg/m<sup>3</sup>, and 27.5% from 20 to 30 5 µg/m<sup>3</sup>. The highest level of PM10 was 190 µg/m<sup>3</sup>. The contribution of heating to dust emission (PM10) was recorded to be 32%, while the contribution of SO<sub>2</sub> was 52%. The contribution of the vehicular traffic to PM10 is 44%, while the contribution to SO<sub>2</sub> is 45%. Lastly, the contribution of industry to PM10 was 19%, while the contribution to SO<sub>2</sub> was estimated to be 16%. Also, SO<sub>2</sub> rised in winter months, while PM10 did not change significantly over the year 2016, and exceeding National Boundary Values were observed to be at the maximum level in February (Gemici 2017; NACACD 2016).

## 2.2. Sample collection and preparation for analyses

For each tree species, the fully matured fresh leaves (~100-150 g) were sampled around tree branches and put into the labelled paper bags in July 2016 and 2017. The leaf samples were mixed to form a representative sample. The leaves samples were specifically collected after the second week of July, so that any increase in air temperatures in July may induce an increase in visible symptoms on the leaves and also facilitates the monitoring of pollution damages. The number of each tree species and mean silvicultural characteristics of the studied tree species are shown in Table 1. Measurements of tree diameter at breast height (DBH) and height were performed on three sample trees for each tree class in summer 2017. The DBH was measured using a diameter tape. Tree age was determined by the dendrochronological approach, coring trees at breast height. Tree heights were measured with a Blume-Leiss clinometer.

The leaf samples were cleaned with distilled water and dehydrated on the blotter papers. The cleaned and dehumidified leaf samples were then air-dried in a sun-free area and ground using a laboratory blender. Later, they were analyzed for their nutrient and radioactivity concentrations in the Central Research Laboratory of Kastamonu University.

## 2.3. Analyses of leaf samples

Gamma-ray spectrometry was performed with FoodGuard-1 3 x 3-inch NaI (TI) model radiation detector (ORTEC, Oak Ridge, USA) in the Central Research Laboratory of Kastamonu University. Firstly, the ground leaves were placed into plastic boxes with a diameter of 8 cm and a height of 8

cm, which were constructed following the geometry of the detector. Secondly, the mouths of the boxes were tightly closed and the boxes were kept for 1 month. Thirdly, the formation of radioactive equilibrium between <sup>238</sup>U and <sup>232</sup>Th and their decay products was provided and the samples were prepared for counting. Finally, the detector was calibrated before the analysis of the samples. To analyze the spectra collected in computer memory, the channel corresponding to input energy must be known. Thus, the types of radioactive nuclei present in the sample can be found. To accomplish the energy calibration, a standard source or resources consisting of nuclei with previously energies was needed. Standard point sources, including the peaks of <sup>109</sup>Cd, <sup>57</sup>Co, <sup>133</sup>Ba, <sup>22</sup>Na, <sup>137</sup>Cs, <sup>54</sup>Mn, and <sup>60</sup>Co, with energies ranged from 80 to 1400 keV were used for the calibration. After the calibration, each sample was counted in the gamma spectrometer for 50000 sec. Activities of radionuclides obtained at end of the measurements were determined by the following equation:

$$\text{Activity} = \frac{\text{Net area}}{\text{Counting time} \times \text{Sample amount} \times \text{Abundance} \times \text{Yield}} \quad (\text{Eq:1})$$

The net areas under the peaks were obtained by subtracting the background from the total area. The radioactivity concentrations of <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K and <sup>137</sup>Cs in the samples were determined by considering the gamma peaks of natural radionuclides, which were the degradation products of these radionuclides. After the determination of the activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K, the activity concentration of <sup>137</sup>Cs isotope in the samples was also determined. The activity concentrations of radionuclides (<sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K, and <sup>137</sup>Cs) were expressed as Bq kg<sup>-1</sup> dry weight.

## 2.4. Statistical analysis

One way ANOVA (Analysis of variance) was applied for analyzing the differences in nutrients, heavy metals and the activity concentration of four radionuclides in the leaf samples of six tree species collected in July 2016 and 2017 from air polluted and unpolluted sites. The statistical analysis was performed using the SPSS program (Version 11 for Windows). Following the results of ANOVAs, Tukey's honestly significant difference (HSD) test ( $\alpha = 0.05$  was used for significance. The relationship between the measured characters was revealed by correlation analysis.

## 3. Result and Discussion

Plants generally take up mineral elements from the soil. Those elements include macronutrients (P, K, S, Mg and Ca) and

micronutrients (Na, Cl, Cr, Mn, Fe, Co, Ni, Cu, Zn, and Mo). The requirement of plants for the macronutrients is quite high concentrations compared to the micronutrients (Dalcars et al., 2014). Final nutrient contents within any plant tissues can vary with nutrient availability in soils, nutrient distribution and redistribution. On the other hand, a number of study have shown that the severity of air pollution, the proximity of plants to pollutants, and also plant species can influence nutrient concentrations in plants (Dalcars et al., 2014; Alaimo and Varrica, 2020).

Mean macronutrient concentrations (ppm) in the leaf samples from the control and polluted sites are shown in Table 2 (Appendix A). For the results of 2016 and 2017, mean Mg concentration in the leaf samples was higher at the polluted sites than at the control sites for all tree species, except for ash tree which showed lower Mg concentration at the polluted sites. Although Mg concentration in ash tree leaves at the polluted sites was not higher than the control sites, among the other tree species ash tree leaves had the highest mean Mg concentration at the polluted sites both in 2016 (6097) and in 2017 (5482), while pine needles had the lowest Mg concentrations (2058 and 1854 respectively). In contrast to mean Mg concentration, mean P concentration in the leaf samples was lower at the polluted sites for maple, pine, plane tree and poplar trees, whereas mean P concentration was higher for cypress and ash trees (Table 2/Appendix A).

At the polluted sites, ash trees had the highest mean P concentration (ppm) in 2016 (2104) and 2017 (2084), whereas poplar trees had the lowest P concentrations (1240 and 1352 respectively). Similar to mean Mg and S concentrations in the leaf samples was higher at the polluted sites for all tree species, exception for ash tree and plane trees. In 2016, pine tree leaves at the polluted sites showed the highest S concentration (5390), while ash tree leaves showed the lowest S concentration (1214). In 2017, however, this time poplar tree leaves had the highest S concentration (7576), while ash tree leaves still had the lowest S concentration (1644). With the exception of maple tree leaves, mean K concentration was also higher at the polluted sites. Plane trees had the highest mean K concentration (25110) both in 2016 and (28620) in 2017 at the polluted sites, while cypress trees had the lowest K concentrations (11120 and 11022 respectively). Mean Ca concentration at the polluted sites was lower for maple, cypress, pine and plane tree leaves, but higher for ash tree and poplar trees. At the polluted sites, maple trees had the highest mean Ca concentration (33680) in 2016 and (28980) in 2017, while pine trees had the lowest Ca concentrations (12390 and 10880, respectively). Mean all macronutrient concentrations in tree leaves from both the control and polluted site generally showed a decrease with time, with the exception of poplar and plane trees, which had a higher concentration in 2017 compared to 2016 (Table 2/Appendix A).

Mean micronutrients concentrations in the leaf samples from the control and polluted sites are shown in Table 2 (Appendix A). In 2016 and 2017 results, mean Na concentration showed lower in the tree leaves of maple, cypress and ash trees at the polluted sites, whereas it was higher in the leaves of pine and poplar tree species. However, plane tree leaves showed lower Na concentrations in 2016, while they had higher Na

concentrations at the polluted sites. Other micronutrients were generally higher at the polluted sites compared to the control sites for all tree species. Only, Co concentration was lower for ash tree and pine tree, and Mn concentration was lower for poplar tree at the polluted site compared to the control site. At the polluted site, the highest amount of Cr, Mn, Fe, Ni and Pb was found in the maple leaves, while the highest Co, Cu and Zn concentration in cypress, pine and poplar leaves respectively in 2016. In 2017, similar results were seen for other micronutrients, but for Zn, the highest concentration was recorded in plane tree leaves. The order of micronutrient concentrations in the tree leaves was Fe, Mn, Zn, Pb, Ni, Cu, Cr and Co for both years (Table 3/Appendix B).

In general, Mg, S and K in the tree leaves increased at the polluted sites, whereas Ca concentration tended to decrease. Considering both years, the order of concentrations was  $Ca > K > Mg > S > P$  at the control site and  $Ca > K > S > Mg > P$  at the polluted site. It has been reported that P, K, and Cl are mobile elements for phloem, and S is variable mobility (Sardans and Penuelas, 2013). The values of essential elements and the amounts of Na and Cl are associated with the high requirements of these elements in plants and also the high transport rates in the phloem (Maillard et al., 2015; Yan et al., 2016). Although Ca is an immobile element, its high content in leaves may be due to the high solubility of this element in the soil. In addition, the fact that the counted elements have a higher requirement in plants than other elements may have caused this result (Dalcars et al., 2014; Kopriva et al., 2019). Results of essential elements are in agreement with the previous studies. Many studies have shown that Ca, K, Mg, P, and S were the most abundant element in the species studied at contaminated and non-contaminated areas. In a study, Alaimo and Varrica (2020) reported that the Ca, K, Mg, P, and S were the most abundant element in the species studied in contaminated and non-contaminated areas. Also, they demonstrated that selecting and using tolerant species with large leaves was suitable for increasing the air quality in industrial areas and urban habitats. Some elements such as K, Na, Mg, P, S, and Cl are easily transferred in the phloem and therefore, they can be found in abundance in the leaves (Kopriva et al., 2019). Since Ca is an important component of cell membranes and cellulose walls, its requirement in plants is quite high. The high amount of Ca in the leaf tissue can be due to the low mobility of this element with the phloem and the limited transfer of Ca accumulated in the leaves to other tissues (Dalcars et al., 2014; Wang et al., 2018). Besides, the accumulation of K was maximum in the leaves of a populus tree at the polluted, and Mg and P in the leaves of maple for both years (Table 2/Appendix A).

Micronutrients or trace elements are essential for the biosynthesis of chlorophyll and secondary metabolites, carbohydrates, and other growth substances. Besides, they are important for the integrity of cellular membranes and stress resistance. However, they are toxic effects at high concentrations, and hence, they are called heavy metals. It has been reported that the ten most important in atmospheric heavy metal pollution arising from heavy traffic, steel and iron industry, mining, fossil fuels, and agricultural activities

are Fe, Al, Pb, Zn, Ti, Mn, Cu, V, Ni, Cr (Przybysz et al., 2019).

The observed values of Cr, Mn, Fe, Co, Ni, Cu, Zn, and Pb concentrations in the leaves of the six trees were in agreement with the previous studies. Baycu et al. (2006) similarly found that metals such as Pb, Fe, Ni, Cd, and Zn increased in an ash tree, poplar and robinia species at the polluted sites (Table 3/Appendix B). Similarly, Dadea et al. (2016) found that Cd, Cu, Mn, Pb, and Zn concentrations in some deciduous plants, such as acer, betula, carpinus, cercis, and robinia growing under the contaminated areas increased compared to the unpolluted sites. Simon et al. (2014) and Samecka et al. (2020) also revealed similar results. Although Fe, Zn and Cu are low mobility and Mn is immobile elements, they are essential minerals in metabolic and physiological processes and they can be found in high amounts in leaf tissue (Maillard et al., 2015). It is well documented that that tolerant genotypes can accumulate more heavy metals in their tissues without any damage and the concentrations of As, I, Sn, and Cr in trees are generally lower than that of Fe, Mn, Al, Zn, Ni, and Cu (Chen et al., 2016; Tzvetkova and Petkova, 2015).

Plants can take both natural and artificial radioactive elements released from nuclear facilities. The man-made radionuclides  $^{90}\text{Sr}$ ,  $^{131}\text{I}$  and  $^{137}\text{Cs}$  are found in the air and soil, but the naturally occurring radioactive nuclides  $^{238}\text{U}$ ,  $^{232}\text{Th}$ , and  $^{40}\text{K}$  are present in the soil (Kumar et al., 2008; Azeez et al., 2019). For this reason, while plants take the first three radionuclides through their leaves and roots from the air and soil, they can only take the three radionuclides through their roots from the soils (Kumar et al. 2008; IAEA 2010).

The activity concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$ , and  $^{40}\text{K}$  in the leaf samples from the control and polluted sites are given in Table 4 (Appendix C). The activity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$ , and  $^{40}\text{K}$  ranged from 14.8 to 92.1 Bq  $\text{kg}^{-1}$ , from 24.5 to 65.1 Bq  $\text{kg}^{-1}$ , from 6.6 to 38.4 Bq  $\text{kg}^{-1}$ , and from 76.88 to 381.3 Bq  $\text{kg}^{-1}$  (the control site in 2016), while the corresponding values in the polluted sites were 7.11-87.3, 26.13-105.8, 6.91-25.33, and 143.11-612.63 Bq  $\text{kg}^{-1}$ , respectively. In 2017, the activity concentration of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$ , and  $^{40}\text{K}$  varied between 40.2-120.8 Bq  $\text{kg}^{-1}$ , between 30.1-138.22, 8.41-43.6 Bq  $\text{kg}^{-1}$ , and 167.3-686.2 Bq  $\text{kg}^{-1}$  in the control sites, while the corresponding values in the polluted sites were 36.8-222.8, 42.48-144.68, 12.62-43.12, and 128.8-714.8 Bq  $\text{kg}^{-1}$ , respectively.

In 2016, the highest radioactivity concentration in the tree leaves was 612.63 Bq  $\text{kg}^{-1}$  ( $^{40}\text{K}$  in ash tree), but it was highest in maple leaves (714.8 Bq  $\text{kg}^{-1}$ ) in 2017. While  $^{232}\text{Th}$  and  $^{40}\text{K}$  activity concentrations generally increased in 2016, there were some increases in  $^{238}\text{U}$  and  $^{232}\text{Th}$  activity concentrations in 2017 (Table 4/Appendix C).

According to the activity concentration changed in the trees, the highest value belonged to the leaves of an ash tree, pine and poplar trees in 2016, but the activity concentration in the trees was generally similar in 2017.  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{137}\text{Cs}$  activity concentrations in the plane leaves were lowest at the polluted sites in both years. While the activity concentration changed in the trees were determined as  $^{40}\text{K} > ^{232}\text{Th} > ^{238}\text{U} > ^{137}\text{Cs}$  in 2016, they were recorded as  $^{40}\text{K} > ^{238}\text{U} > ^{232}\text{Th} > ^{137}\text{Cs}$  in 2017 (Table 4/Appendix C).

As seen in Table 4 (Appendix C), among the measured radionuclides,  $^{40}\text{K}$  was seen to accumulate in the most abundant in the tested trees. The high levels of  $^{40}\text{K}$  and  $^{238}\text{U}$  in the tree leaves were associated with the ecological and physiological behaviour of both elements. Uranium is analogous to Ca, which is an essential element in plants, and  $^{40}\text{K}$  is analogous to K, and therefore its activity in plant tissues is high (Buysse et al., 1995; Kumar et al., 2008). Plants take  $^{40}\text{K}$  in the same manner as the essential element of K and use it in similar functions, and therefore its amount in vegetative tissue is high. Possible reasons to increase in the activity concentration of  $^{40}\text{K}$  in trees include the fact that the activity concentrations in the soils and solubility in water of  $^{40}\text{K}$  are much higher than those of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$ , and  $^{40}\text{K}$  is also a light element (Kumar et al., 2008; Azeez et al., 2019). Additionally, the upper activity of  $^{40}\text{K}$  might be attributed to the higher biological requirement of plants for potassium. In addition, some species can accumulate  $^{238}\text{U}$  at a higher extent, especially in older leaves, and are therefore considered bioindicators. Results are consistent with the previous results. Yoshihara et al. (2014) studied with the foliar parts of 10 woody species to assess the radiocesium concentrations between 2011-2013 and results indicated that the mean concentrations in 2011, 2012, and 2013 for the evergreen species was 1440 Bq  $\text{kg}^{-1}$ , 790 Bq  $\text{kg}^{-1}$ , and 320 Bq  $\text{kg}^{-1}$ , respectively, whereas that for the deciduous species was 320 Bq  $\text{kg}^{-1}$ , 290 Bq  $\text{kg}^{-1}$ , and 60 Bq  $\text{kg}^{-1}$ , respectively, which were much higher than the values we found. Cengiz and Çağlar (2019) revealed that the radioactivity concentrations were varied in the range of  $19.74 \pm 3.7$  to  $85.01 \pm 5.6$  Bq  $\text{kg}^{-1}$  for  $^{232}\text{Th}$ , and from  $795.82 \pm 9.8$  to  $1056.28 \pm 14.4$  Bq  $\text{kg}^{-1}$  for  $^{40}\text{K}$  in some medicinal and herbal species.

#### 4. Conclusion

In this study, it has been shown that the accumulation capacity of macro-and micro-nutrients and radionuclides in the tree leaves varies according to tree species, element type as well as times. Cypress and poplar tree species had higher macronutrients (K, S, P, Ca) in the leaves at the polluted sites than at the unpolluted sites. When both years are evaluated together, the order of the elements is Ca, K, Mg, S and P for the control sites, while it is Ca, K, S, Mg and P for the polluted sites. On the other hand, micronutrients in the leaf samples are higher at the polluted sites for both years. The order of the micronutrients is Fe, Mn, Zn, Pb, Ni, Cu, Cr and Co for both years. While the activity concentration changes in the trees are recorded as  $^{40}\text{K} > ^{232}\text{Th} > ^{238}\text{U} > ^{137}\text{Cs}$  in 2016, they are recorded as  $^{40}\text{K} > ^{238}\text{U} > ^{232}\text{Th} > ^{137}\text{Cs}$  in 2017. The highest concentration of radioactivity at the polluted sites in 2016 was for ash trees and pine trees, but in 2017 the concentration of radioactivity in the tree leaves was mostly similar except for plane trees. Considering both years, the plane tree had the lowest activity concentration. Taking all data into an account, the order of the macronutrient and micronutrient accumulation capacity and the activity concentration of radionuclides in the tree leaves at the polluted sites are poplar > ash tree > maple > pine > plane tree = cypress for 2016, while the order is poplar > cypress = maple > ash tree > pine = plane tree for 2017. As a result, the lowest value in terms of the investigated parameters is noted in the leaves of the plane

tree and pine trees, but the plane tree has the highest value in terms of radioactive elements. It can be concluded that poplar, ash tree, and maple could be recommended for future biomonitoring studies in order to improve air quality in urban areas. The best part of this study is to be the first study to investigate the radioactivity concentrations in the forest trees in the Kastamonu region. The results of the present study are useful in the assessment of the exposures of radiation doses and would be a valuable database for future estimations of the impact of radioactive pollution.

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## Appendix A

Table 2. Variation in macro nutrients (Mg, P, S, K, and Ca concentrations-ppm) in the leaf samples of six tree species collected from the polluted and unpolluted sites.

Tree species	Study sites	Mg		P		S		K		Ca	
		2016	2017	2016	2017	2016	2017	2016	2017	2016	2017
Maple	Control	4224±32d*	4674±32d	2656±4d	2476±4d	2902±3c	3246±3c	20050±30d	24640±30d	36500±30e	32300±30d
	Polluted	5683±36e	5213±36e	1617±3b	1834±3b	3863±4d	3457±4c	15330±20c	19630±20c	33680±30e	28980±30c
Cypress	Control	1101±17a	986±17a	1454±3a	1246±3a	1404±2a	1326±2a	10430±20a	9530±20a	34130±30e	26130±30c
	Polluted	3302±28c	2402±28c	1703±3b	1528±3a	2069±3b	1852±3b	11120±20a	11022±20a	28850±30d	22450±30b
Ash tree	Control	7022±41f	6455±41f	2075±4c	1926±4b	2954±3c	2462±3b	12780±20b	11480±20a	27780±30d	24660±30b
	Polluted	6097±42e	5482±42e	2104±4c	2084±4c	1214±10a	1644±10a	15480±20c	14680±20b	32940±30d	29850±30c
Pine	Control	1226±18a	1024±18a	2614±5d	2212±5c	2248±3b	2124±3b	12740±20	11440±20a	16540±20b	14260±20a
	Polluted	2058±24b	1854±24b	1543±3a	1352±3a	5390±5f	4180±5d	15990±20c	14650±20b	12390±20a	10880±22a
Plane tree	Control	4656±33d	4436±33d	1647±3b	2014±3c	5283±5f	4822±5e	20450±30d	24260±30d	36350±30e	38650±30e
	Polluted	5356±35	5128±35e	1692±3b	1744±3b	3375±4c	3026±4c	25110±30e	28620±30e	25470±30c	28480±30c
Poplar	Control	1485±26a	1885±26b	1614±4b	1802±4b	3364±4c	3956±4d	10610±20a	12610±20a	16050±30b	26050±30c
	Polluted	3414±36c	4414±36d	1240±3a	1430±3a	4576±6e	7576f±6	14300±20c	16200±20b	22660±30c	31660±30d
F		158150.5	158150.5	1321375.4	1321375.4	2.055	2.055	96218955.4	96218955.4	148352002.5	148352002.5
Sig.		<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001

\* Means indicated with different letters within same column are significantly different (P < 0.05)

## Appendix B

Table 3. Variation in micro nutrients (Na, Cl, Cu, Zn, Pb, Cr, Mn, Fe, Co and Ni concentrations-ppm) in the leaf samples of six tree species collected from the polluted and unpolluted sites.

Tree species	Study sites	Na		Cl		Cu		Zn		Pb	
		2016	2017	2016	2017	2016	2017	2016	2017	2016	2017
Maple	Control	1020±160c*	1350±160c	951±3.0b	1420±3.0c	9.44±0.3b	9.60±0.3a	25.0±0.3b	22.0±0.3a	1.4±0.1a	1.42±0.1a
	Polluted	920±170c	1055±170b	2250±4.0d	1856±4.0d	14.10±0.3	16.20±0.3b	68.5±0.6d	82.0±0.6b	22.4±0.3	24.60±0.3d
Cypress	Control	1810±190f	1510±190c	2039±5.0c	2168±5.0d	7.30±0.3a	8.40±0.3a	18.4±0.2a	22.4±0.2a	2.2±0.1a	2.42±0.1a
	Polluted	1670±170e	1420±170c	1828±3.0c	2346±2.0d	11.20±0.3b	14.80±0.3b	47.6±0.5c	82.2±0.5b	15.7±0.3c	15.88±0.3c
Ash tree	Control	1090±180c	1245±180b	2093±4c	2226±4.0d	11.70±0.3b	12.14±0.3a	107.0±0.5e	85.4±0.5b	2.3±0.2	2.83±0.2a
	Polluted	570±210b	944±210b	2402±3e	2562±3.0e	14.60±0.3c	17.80±0.3c	125.7±0.3	89.4±0.3b	7.9±0.2b	8.60±0.2b
Pine	Control	1260±180d	1056±180b	623.1±1.8a	716.1±1.8a	11.50±0.3b	12.45±0.3a	56.8±0.4c	76.8±0.4b	2.1±0.2a	2.42±0.2a
	Polluted	1480±180e	1268±180b	686.2±1.5a	826.2±1.5a	16.00±0.3c	18.80±0.3c	82.3±0.4d	102.4±0.4c	11.1±0.2	11.60±0.2c
Plane tree	Control	1890±220f	2990±220d	5189±7.0f	5355±7.0g	8.10±0.3a	8.60±0.3a	29.8±0.8ab	186.2±0.8d	2.1±0.2a	2.34±0.2a
	Polluted	880±190c	3470±190e	5543±5.0g	4360±5.0f	8.10±0.3a	9.20±0.3a	37.7±0.2c	194.6±0.2d	5.2±0.2b	5.64±0.2b
Poplar	Control	360±160a	628±160a	825.97±3.2b	944±3.20b	7.30±0.3a	7.80±0.3a	74.3±0.4d	66.3±0.4b	2.5±0.2a	2.90±0.2a
	Polluted	1090±170c	1450±170c	2091±11.0c	2216±11.0d	11.3±0.3b	15.40±0.3b	148.4±0.6f	124.2±0.6c	5.4±0.2b	6.50±0.2b
F		1661674.3	1661674.3	21219104.4	21219104.4	2238.2	2238.2	81546.4	81546.4	52.1	52.1
Sig.		<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001

\* Means indicated with different letters within same column are significantly different (P < 0.05)

## Appendix B

Table 3. Continued

Tree species	Study sites	Cr		Mn		Fe		Co		Ni	
		2016	2017	2016	2017	2016	2017	2016	2017	2016	2017
Maple	Control	3.5 ±0.1a	3.53 ±0.1a	129±0.7d	186±0.7d	340.8±1.7b	260.8±1.7b	4.8 ±0.9a	4.66 ±0.9a	15.5±0.3b	17.40±0.3c
	Polluted	14.5 ±0.3d	16.85 ±0.3c	565.2±1.4i	432±1.4g	5017±8.0i	5622±8h	8.1 ±1.3a	10.40 ±1.3b	19.8±0.3c	22.40±0.3
Cypress	Control	4.6 ±0.1a	4.9 ±0.1a	41.7±0.4a	36.7±0.4a	516.4±2.0e	816.4±2d	7.8 ±1.3a	7.58 ±1.3.a	9.1±0.3a	8.80±0.3a
	Polluted	9.4 ±0.2c	9.8 ±0.2b	337.9±1h	424.8±1g	1246.0±10.0	1345±10	10.2 ±1.4b	12.45 ±1.4b	12.7±0.3b	12.30±0.3b
Ash tree	Control	3.9 ±0.1a	3.6 ±0.1a	68.9±0,5	84.9±0.5b	494.3±2.0d	288.3±2b	7.0 ±1.2a	7.80 ±1.2a	10.5±0.3a	11.45±0.3a
	Polluted	4.5 ±0.1a	4.8 ±0.1a	140.7±0.7e	165.4±0.7d	1896±4.0h	2034±4f	6.4 ±1.2a	6.25 ±1.2a	17.9±0.3b	18.11±0.3c
Pine	Control	4.5 ±0.1a	5.22 ±0.1a	77±0.5b	88±0.5b	161.3±1.1a	178.3±1.1a	8.0 ±1.3a	8.55 ±1.3	7.8±0.3a	7.25±0.3a
	Polluted	7.3 ±0.2b	8.6 ±0.2b	149.7±0.7e	178.4±0.7d	2161±4.0i	2196±4f	6.3 ±1.2a	6.22 ±1.2a	10±0.3a	11.30±0.3a
Plane tree	Control	5.0 ±0.1a	5.7 ±0,1	96.7±0.6c	144±0.6c	488.5±2.0c	548±2.0c	7.8 ±1.3	7.66 ±1.3	14.7±0.3b	15.40±0.3b
	Polluted	7.4 ±0.2b	7.3 ±0.2b	211±0.9g	316±0.9f	1228±5.0g	2344±5g	8.4 ±1.3a	9.20 ±1.3b	18.8±0.3	19.20±0.3c
Poplar	Control	3.4 ±0.1a	4.2 ±0.1a	194.4±0.8f	272±0.8e	732.3±2.5f	814±2.5d	4.9 ±1a	4.60 ±1.0a	10.1±0.3a	10.88±0.3a
	Polluted	3.8 ±0.1a	5.2 ±0.1a	128.8±0.6d	186±0.6d	1019±3.0g	1104±3.0e	7.1 ±1.3a	7.80 ±1.3a	16.7±0.3b	14.88±0.3b
F		2549.4	2549.4	299680.7	299680.7	17568615.3	17568615.3	388.4	388.4	66.5	66.5
Sig.		<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001

\* Means indicated with different letters within same column are significantly different (P < 0.05)

## Appendix C

Table 4. Variation of  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{137}\text{Cs}$ , and  $^{40}\text{K}$  in the leaf samples of six tree species collected from polluted and unpolluted sites ( $\text{Bq kg}^{-1}$ ).

Tree species	Study sites	$^{238}\text{U}$		$^{232}\text{Th}$		$^{137}\text{Cs}$		$^{40}\text{K}$	
		2016	2017	2016	2017	2016	2017	2016	2017
Maple	Control	26.32±4.2c	56.4±7.3c	25.6±3.6a	41.4±5.3b	38.4±3.5d	29.13±3.5d	381.3±42.8e	581.11±62.40g
	Polluted	49.66±7.4e	95.9±9.4e	41.9±6.8c	48.7±6.8c	11.3±1.2b	16.1±1.6b	289.9±34.7d	714.8±75.42i
Cypress	Control	46.82±5.7e	40.2±6.2b	30.1±4.5a	30.1±4.2a	36.3±3.7d	43.6±6.8e	159.8±25.2b	167.3±21.12c
	Polluted	45.00±5.4e	108.4±9.5e	51.8±8.4d	91.5±8.5f	13.5±1.3b	21.47±3.4c	155.5±21.12b	236.2±26.34d
Ash tree	Control	27.78±2.6c	68.10±7.5d	24.5±.4a	47.5±6.4c	6.6±0.44a	8.41±2.2a	334.2±37.2d	378.0±32.14e
	Polluted	40.9±6.8d	191.88±18.4g	51.28±8.3d	114.6±10.6	6.88±0.6a	31.2±3.6d	612.63±70.4g	128.8±16.46a
Pine	Control	92.1±9.2g	44.35±4.7b	32.32±4.2b	64.18±7.4d	12.58±1.2b	26.1±2.8c	76.88±.8.9a	399.18±36.52f
	Polluted	87.3±8.4g	69.86±7.5d	76.51±12.6f	79.68±8.4e	25.22±2.74c	29.12±3.1d	143.11±10.6b	145.27±10.5b
Plane tree	Control	65.8±7.6f	65.76±8.3d	65.1±11.4e	65.13±7.2d	33.8±3.2	33.82±4.6d	182.59±16.7c	182.58±13.6c
	Polluted	33.73±3.8c	36.8±4.7a	26.13±3.2a	42.48±4.8b	6.91±0.2a	12.62±1.6b	204.48±20.54c	348.22±34.9e
Poplar	Control	14.8±1.7b	120.8±12.5f	41.9±5.3c	138.22±14.12g	24.3±2.6c	23.42±3.7c	182.8±16.42c	686.2±66.8h
	Polluted	7.11±0.4a	222.8±18.22h	105.8±13.8g	144.68±15.6h	24.2±3.2c	43.12±6.2e	501.5±65.44f	680.3±64.2h
F		561673.8		35638757.3		7537714.2		7265852.2	
Sig.		<0.001		<0.001		<0.001		<0.001	

\* Means indicated with different letters within same column are significantly different ( $P < 0.05$ )