ATMOSPHERIC DEPOSITION IN MATURE FIR (*Abies nordmanniana*) STANDS IN ALADAĞI-BOLU, TURKEY

KINIŞ, S.[1] – YILDIZ, O.[2*]

[1]*General Directorate of Forestry, Bolu, Turkey*

[2]*Düzce University, Forestry Faculty, Düzce, Turkey*

[*]*Corresponding author*
e-mail: oktayyildiz@duzce.edu.tr

(Received 30th Apr 2020; accepted 11th Aug 2020)

Abstract. The aim of this study was to determine the atmospheric deposition in fir forests (*Abies nordmannianna* (Steven)) in the Bolu Aladağ region. The study area selected was located in the mountainous Aladağ region stretching along the south of Bolu Province in the Western Black Sea Region of Turkey. Four elevation zones (1000, 1200, 1400, and 1600 m above sea level) were designated on the closed-canopy fir forest-dominated northern slopes of the mountain. Sampling plots were established for each elevation zone, eight on randomly located sites under the canopy and three on adjacent open sites. For each elevation zone, open site, throughfall, and stemflow deposition samples were collected between April and November for two years (2013-2014). Chemical analysis of the samples was performed in the ICP laboratory located within the Aegean Forestry Research Institute in İzmir. One-way ANOVA was carried out for each variable to determine the differences in the sampling plots. Sulfate, nitrogen, potassium, and calcium were the main ions deposited in the fir forests in the 1000-1600-m elevation zones in Aladağ. The acid neutralization capacity of the deposition samples collected in the open site plots was high for all elevation zones.
Keywords: *atmospheric pollution, fir forest, Level II site, ICP-Forests, Turkey*

Introduction

Atmospheric pollution caused by intense agricultural practices, industrial activities, and increased use of fossil fuel has become one of the main agents disrupting forest ecosystems. The challenge of environmental problems goes beyond the jurisdiction of states, and their solution requires international cooperation. From the 1960s onward, international efforts for environmental protection have been initiated, with the UN in particular playing a major role in organizing international efforts. The 1979 Convention on Long-Range Transboundary Air Pollution and its seven protocols have emphasized international cooperation as a solution. As stated in the Rio Declaration Principle 10: "Environmental issues are best handled with the participation of all concerned parties."

After the increase in forest losses beginning from the second half of the 20th century until the 1980s, a number of studies were initiated to investigate the impact of air pollution on European forests (Bussotti and Ferretti, 1998; Erisman et al., 2008; Novotný et al., 2016; Kosonen et al., 2019; Schmitz et al., 2019; Thimonier et al., 2019; Etzold et al., 2020). A network was established in the early 1980s to monitor and evaluate the status of forests in Europe (Augustin et al., 2005). Most of these studies concentrated on spruce, fir, and beech forests in central and northern Europe. The International Cooperation Program for the Monitoring and Evaluation of the Impact of Air Pollution on Forests (ICP-Forests) was launched in 1985 within the context of the United Nations Economic Commission for Europe (UNECE) Decision on Long-Range Transboundary Air Pollution. Currently, 42 countries in and around Europe are included in the program

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

(Anonymous, 2010). Within the scope of the study, 800 intensive monitoring plots were established and atmospheric deposition data were collected in 540 of these plots. Turkey joined the study in 2007 and established monitoring plots in main forest types across the country. In this context, Turkey has established 54 Level II intensive monitoring plots. One of the subjects studied in the intensive observation sites is atmospheric deposition. Most of the studies dealing with air pollution effects on forests have been conducted in central and northern Europe. Data from other parts of Europe, the Balkans, and surrounding areas including Turkey are limited. Therefore, the aim of the current study was to determine the atmospheric deposition in fir forests [*Abies nordmannianna* (Steven) Spach subsp. *Equi-trojani* (Asch. & Sint. ex Boiss.) Coode & Cullen] located in the Bolu Aladağ region where one of the ICP-Level II plots was established (Anonymous, 2015). Thus, national data from these fir stands will contribute to the evaluation of the forest conditions affected by transboundary air pollution.

Materials and Methods

Study area

The study area was selected in the mountainous Aladağ region that stretches along the south of Bolu Province in the Western Black Sea Region of Turkey. The geographical position of the area lies between 30º 32’ – 32º 36’ east longitudes and 40º 06’ – 41º 01’ 40º 06 ' 41º 01' north latitudes. The mean annual temperature of the area is 13 °C and it receives more than 800 mm average annual precipitation (Kantarcı, 1980; Şahin, 2017). The region has a mild, semi-humid maritime climate generated from the Black Sea on the north and under the influence of the Central Anatolian steppe climate from the south. The prevailing winds in the area are in the north, north-east, and north-west directions. The fir zone above 1300-m elevation is characterized by fog; therefore, a more humid and cooler climate prevails above this elevation (Kantarcı, 1980). The Bolu meteorological station is the only meteorological station in the vicinity of the sampling sites, which are located about 20 km to the south at about 900 m above sea level (asl) (*Fig. 1*).

The vegetation on the northern slope of Aladağ is primarily composed of +100-year-old Turkish fir trees. Oak and beech species are mixed in the canopy cover in some locations. The soil texture ranges from loamy-clay to clay-loam and rarely, sandy loam. The soil is relatively deep, well drained, and derived from basaltic-andesite material from the volcanic and pyroclastic rocks of the lower and middle Miocene period. Soil in the region has been classified as gray-brown forest soil according to the old soil classification. Fir stands, covering about 670 thousand ha, compose almost 3% of the forests in Turkey (Mayer and Aksoy, 1998). Fir species have established forests in moist vegetation zones 650-2000 m asl in the northwest part of Anatolia (Kantarcı, 1980). The fir zone in Aladağ stretches for 1000 m along the north-facing slope of the mountain at an elevation of 1660 m. The south-facing slope is dominated by Scots pine (*P. sylvestris*) forests.

Methods

Four elevation zones (1000, 1200, 1400, and 1600 m asl) were designated on the northern slopes of the mountain dominated by closed-canopy fir forests with leaf area index between 3 and 5. At each elevation zone, sampling plots 20 × 30 m in size were established under the forest canopy and on nearby open sites, following the ICP-Forests

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

Manual guidelines. The sampling sites were located near a Level II ICP-Forests site established for Turkish fir stands. For each elevation zone, open site, throughfall, and stemflow deposition samples were collected from April through November for two years (2013 and 2014). Throughfall and stemflow data were subtracted from the total precipitation collected on the open site plots to calculate the interception rates. Samples were not collected between December and March because roads were closed due to snow. Based on 60 years of climatic data gathered from the Bolu station, about 39% of the precipitation occurs between December and March. In this study, since precipitation was sampled between April and November, we were able to calculate the total annual precipitation by increasing the collected precipitation data by about 39%. Based on these calculations, the sampling sites at 1000-1200 m altitudes received about 75% higher precipitation than the meteorological station average for 900 m asl.

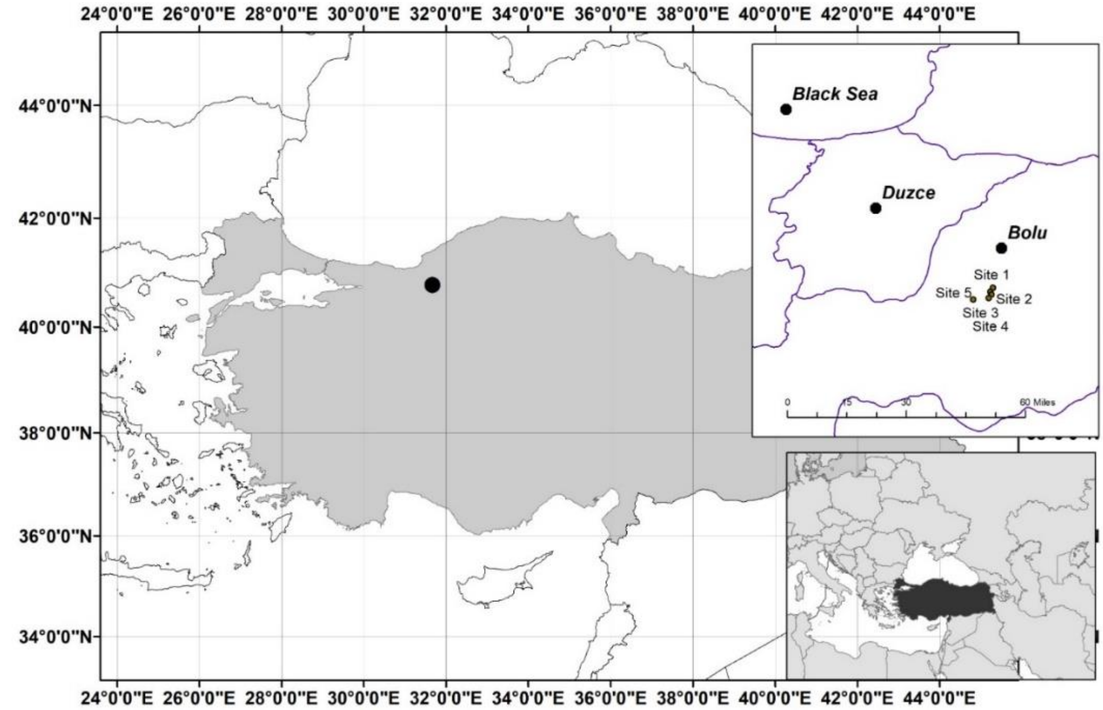

Figure 1. *Location of sampling sites for deposition study in Aladağ fir forest*

For each elevation zone, sampling plots were established, eight on randomly located spots under the canopy and three on adjacent open sites (20-50 m distant). Deposition samples were collected using 500-mL polyethylene (PE) bottles connected to funnels having a 100-cm^2 opening. All sample containers were numbered and placed on top of a PVC pole 1 m above ground. In order to collect stemflow, on each sampling plot, three Grade-3 (diameter class) fir trees were chosen to represent the stand, and spiral collectors were installed on the tree stems at a height of 0.5-1.5 m from the ground (*Fig. 2*).

The collectors were tightly attached to the stem to prevent leakage and containers were placed near the tree. The deposition samples were collected weekly and processed according to Part XIV of the ICP-Forests Manual: “Sampling and Analysis of Deposition, Methods and Criteria for Harmonized Sampling”. The average sample at each collection was formed by proportional mixing of samples from each collector in

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

order to represent the variability. Each week, the field was visited and samples were collected, placed in boxes containing ice batteries, brought to the laboratory, and stored at 4 °C until they were analyzed.

***Figure 2.** Throughfall, open site and stemflow precipitation collection in Aladağ fir forest*

Laboratory analysis

Chemical analyses of the samples were made in the ICP laboratory established within the Aegean Forestry Research Institute in İzmir. The samples were filtered through a 0.45-µm pore membrane before analysis. First, the solution pH (Thermo Scientific™ Orion™ 3-Star Benchtop pH Meter) and electrical conductivity (EC) were measured, and then the ion, anion, and cation concentrations of the samples were analyzed via liquid chromatography (Thermo Scientific Dionex™ ICS-5000+ DC). The mandatory variables required by the ICP-Forests Manual for Level-II sites were analyzed for each sample. For this, each deposition sample was analyzed for fluorine (F^-), chlorine (Cl^-), nitrite (NO_2^-), bromine (Br^-), phosphate (PO_4^{-3}), sulfate (SO_4^{-2}), nitrate (NO_3^-), lithium (Li^+), sodium (Na^+), potassium (K^+), calcium (Ca^{+2}), magnesium (Mg^{+2}), and ammonium (NH_4^+) concentrations.

Statistical analysis

For each variable, one-way analysis of variance (ANOVA) was performed to determine the differences in the sampling plots. The Tukey mean separation test was used for variables where the ANOVA results differed. The non-parametric Kruskal-Wallis test was applied to the total amount of PO_4^{-3}, Cl^-, and NH_4^+ deposition in the open sites and the monthly stemflow NO_2^- and Na^+ data because they did not show normal distribution. The SAS (1996) package program was used in the analysis, with results considered to be different at the level of $\alpha = 0.05$.

Results

The sampling plots at 1400 and 1600 m received about 17% more rainfall than those of the sites at 1000 and 1200 m during the 2013-2014 sampling period. For each sampling site, about 30% of the total precipitation was intercepted by the canopy crown. Stemflow accounted for about 1% of the precipitation (*Fig. 3*).

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

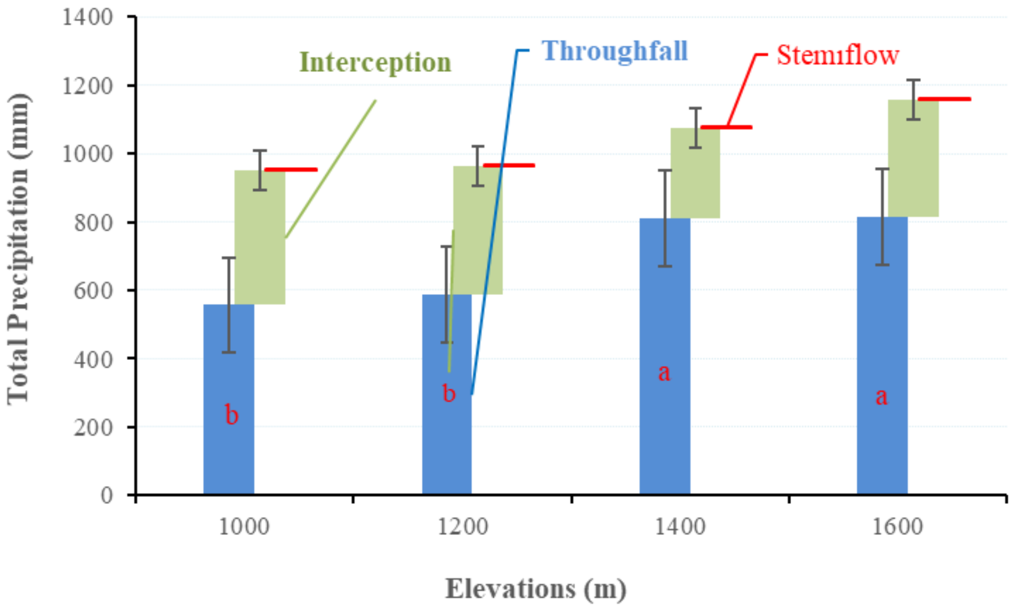

Figure 3. *Means ± standard deviation of annual precipitation (mm) in Aladağ fir forest (means followed with the same letter are not significantly different at α = 0.05*

Since the EC values across the sampling sites were below 0.1 dS m^{-1}, there was no salinity problem in the depositions. The pH values of the deposition samples averaged 6 and were not significantly different among sampling sites.

Anions

Nitrate concentration in both the open site and throughfall depositions were reduced by about 15% and 75%, respectively, above the 1000-m elevation zone. Throughfall NO_3^- concentrations were 600% and 200% greater than in open site samples at 1000 m and other sites, respectively. Sulfate concentrations did not show significant differences among elevation zones. However, SO_4^{-2} concentrations in throughfall samples were 300% higher compared to those of open sites. Stemflow SO_4^{-2} at 1000-m elevation was 200% higher than at the other elevation zones. Phosphate concentrations in both open site and throughfall samples showed significant variation within each sampling plot. Open site Cl^- concentrations were not significantly different among elevation zones; however, they increased 200-300% in throughfall samples compared to those of open sites across all sampling sites. Stemflow Cl^- concentration at 1600 m was 65%, which was 40% lower than at 1000 m and the averages of the 1200 and 1400 m elevation zones. Open site NO_2^- concentration decreased by 75% at 1600 m compared to the other elevation zones. Throughfall NO_2^- concentrations were 600%, 200%, and 1200% higher than those of open sites at 1000-m, 1400-m, and 1600-m elevation zones, respectively. Neither open site nor throughfall F^- concentrations changed significantly among the elevation zones. However, throughfall F^- concentrations were about 250% higher compared to the adjacent open sites for each elevation zone. Stemflow F^- concentration at 1200- and 1400-m elevation zones decreased to half of the values recorded at 1000 m, and decreased again by 50% at 1600 m compared to the previous elevation. Open site, throughfall, and stemflow Br^- concentrations showed significant variation within each sampling plot (*Table 1*).

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

***Table 1.** Means (mg l^{-1}) ± standard deviation of anion concentrations of throughfall, stemflow, and open site bulk depositions collected at different elevations (m) in Aladağ fir forest*

Elevation (m)		1000	1200	1400	1600
NO_3^-	Throughfall	6.9 ± 3.5aA	1.8 ± 1.01bA	1.6 ± 1.1bA	1.7 ± 1.06bA
	Open site	1.1 ± 0.8aB	0.87 ± 0.6bA	0.79 ± 0.49bA	0.84 ± 0.73bA
	Stem flow	0.7 ± 0.2aB	0.74 ± 0.25aA	0.99 ± 0.35aA	0.27 ± 0.25aA
SO_4^{-2}	Throughfall	13 ± 9aB	13.8 ± 8.8aB	10 ± 8.1aB	14.9 ± 8.7aB
	Open site	3.9 ± 3.1aC	4.3 ± 3.5aC	3.5 ± 2.9aC	3.6 ± 3.2aC
	Stem flow	169 ± 2aA	83 ± 8bA	89 ± 1.1bA	94 ± 1.5bA
PO_4^{-3}	Throughfall	0.6 ± 0.5aA	1.51 ± 1.14aA	0.38 ± 0.21aA	0.07± 0.02aA
	Open site	0.17 ± 0.07aA	0.27 ± 0.23aA	0.08 ± 0.05aA	0.05± 0.02aA
	Stem flow	17 ± 14.5aA	4.1 ± 3.4aA	1.8 ± 1.5aA	2.6 ± 1.8a
Cl^-	Throughfall	3.2 ± 3a	3 ± 2a	2.1 ± 1.1a	2.8 ± 1.9a
	Open site	0.9 ± 0.7a	0.9 ± 0.7a	1 ± 0.89a	0.9 ± 0.8a
	Stem flow	33 ±17aA	21± 13bA	20 ± 16bA	12 ± 10cA
NO_2^-	Throughfall	0.43 ± 0.27aA	0.19 ± 0.14bA	0.15 ± 0.11bA	0.25± 0.16bA
	Open site	0.07 ± 0.004aB	0.11 ± 0.09aA	0.08 ± 0.03aB	0.02 ± 0.01bB
	Stem flow	0.29 ± 0.14aA	0.09 ± 0.05aA	0.27 ± 0.19aA	0.06 ± 0.04aB
F^-	Throughfall	0.13 ± 0.11aA	0.14 ± 0.12aA	0.11 ± 0.08aA	0.11± 0.06aA
	Open site	0.05 ± 0.037aB	0.057 ± 0.036aB	0.044 ± 0.035aB	0.04± 0.036aB
	Stem flow	0.95 ± 0.79aA	0.44 ± 0.39aA	0.46 ± 0.36aA	0.26 ± 0.21aA
Br^-	Throughfall	0.002 ± 0.0004aA	0.006 ± 0.002aA	0.002 ± 0.0009aA	0.003 ± 0.001aA
	Open site	0.003 ± 0.0026aA	0.001 ± 0.0002aA	0.001 ± 0.0008aA	0.003 ± 0.001aA
	Stem flow	0.014 ± 0.012aA	0.078 ± 0.054aA	0.073 ± 0.071aA	0.04 ± 0.03aA

For each row, variable means followed by the same lower-case letter are not significantly different at $\alpha = 0.05$. For each column, variable means followed by an upper-case letter are not significantly different at $\alpha = 0.05$

For stemflow, no correlation was detected between anion concentrations and tree diameter.

Cations

None of the open site, throughfall, or stemflow K^+ concentrations were significantly different among elevation zones. However, throughfall K^+ concentrations were 300-500% higher than those of open site plots across the elevation zones. Stemflow K^+ concentrations were 400–700% higher than those of throughfall at each elevation zone. Open site NH_4^+ concentrations at 1200 m were 250% higher than those at 1000-m elevation, but this value decreased by 75% at the 1400- and 1600-m elevation zones compared to values at 1200 m. However, throughfall and stemflow NH_4^+ concentrations were not significantly different from those of open site plots at any of the elevation zones. Neither open site nor throughfall Ca^{+2} concentrations were significantly different among elevation zones. However, throughfall Ca^{+2} concentrations at 1000-m elevation zones were 150% higher compared to the adjacent open site plots. Stemflow Ca^{+2} concentrations at 1600 m decreased to 50% of those at other elevation zones. Stemflow Ca^{+2} concentrations were about 500% higher than those of the open site plots at each elevation zone. Neither open site nor throughfall Na^+ concentrations varied significantly among elevation zones. However, throughfall Na^+ concentrations were about 150% higher than

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

those of adjacent open site plots at 1000-m elevation zones. Stemflow Na^+ concentrations were about 400% higher than those of throughfall at each elevation zone. Stemflow Na^+ concentrations at 1600 m decreased by about 30% compared to the other elevation zones.

Neither open site nor throughfall Mg^{+2} concentrations significantly differed among elevation zones. However, stemflow Mg^{+2} concentrations at 1600 m decreased by about 50% compared to those at the other elevation zones. Throughfall Mg^{+2} concentrations were about 250% higher than those of the adjacent open site plots at each elevation zone. Stemflow Mg^{+2} concentrations were 300%–1000% higher than those of throughfall at each elevation zone. Neither open site nor throughfall Li^+ concentrations showed significant differences among elevation zones. Throughfall Li^+ concentrations were also similar to those of the adjacent open sites at each elevation zone; however, stemflow Li^+ concentrations at 1200-, 1400-, and 1600-m elevation zones were almost 400% higher than open site and throughfall concentrations (*Table 2*).

Table 2. *Means (mg l^{-1}) ± standard deviation of cation concentrations of throughfall, stemflow, and open site bulk depositions collected at different elevations (m) in Aladağ fir forest*

Elevation (m)		**1000**	**1200**	**1400**	**1600**
K^+	Throughfall	10 ± 6.3aB	15 ± 9.6aB	8.5 ± 4.8aB	8.9 ± 3.2aB
	Open site	2.1 ± 1.2aC	2.1 ± 1aC	2.9 ± 2.5aC	1.9 ± 0.9aC
	Stemflow	76.5 ± 55aA	61 ± 45aA	58 ± 43aA	42 ± 23.19aA
NH_4^+	Throughfall	1.3 ± 1.01aA	2.1 ± 1.8aA	1 ± 0.7aA	0.86 ± 0.71aA
	Open site	1.13 ± 1.01bA	2.7 ± 1.9aA	0.9 ± 0.8bA	0.97 ± 0.78bA
	Stemflow	3.18 ± 3.09aA	2.2 ± 2aA	2.8 ± 2.5aA	3.1 ± 2.7aA
Ca^{+2}	Throughfall	6.9 ± 4.6aB	6.4 ± 3.7aB	5.2 ± 3.5aB	5.7 ± 2.7aB
	Open site	3.4 ± 2.8aC	3.9 ± 3.2aB	3.2 ± 2.1aB	3.1 ± 1.9aB
	Stemflow	37 ± 20aA	27 ± 20aA	33 ± 20aA	17 ± 13bA
Na^+	Throughfall	1.6 ± 1.1aB	1.56 ± 1.4aB	1.2 ± 0.9aB	1.4 ± 1.1aB
	Open site	0.7 ± 0.5aC	1 ± 0.76aB	0.7 ± 0.5aB	0.68 ± 0.47aB
	Stemflow	6.2 ± 4.3aA	6.1 ± 5.5aA	7.3 ± 5.9aA	3.9 ± 3.8bA
Mg^{+2}	Throughfall	1.14 ± 0.91aB	1.2 ± 0.9aB	0.8 ± 0.5aB	1 ± 0.8aB
	Open site	0.32 ± 0.19aB	0.4 ± 0.3aB	0.4 ± 0.3aB	0.26 ± 0.19aB
	Stemflow	8.6 ± 5.4aA	6 ± 4.3aA	7.3 ± 5aA	3.6 ± 2.5bA
Li^+	Throughfall	0.001 ± 0.001aA	0.001 ±0.001 aB	0.001 ± 0.001aB	0.001 ± 0.001aB
	Open site	0.0036 ±0.001 aA	0.0010 ± 0.001aB	0.0010 ± 0.001aB	0.003 ±0.001 aB
	Stemflow	0.0014 ± 0.001bA	0.0057 ± 0.001aA	0.0044 ± 0.001aA	0.005 ± 0.001aA

For each row, variable means followed by the same lower-case letter are not significantly different at α = 0.05. For each column, variable means followed by an uppercase-letter are not significantly different at α = 0.05

For stemflow, no correlation was detected between cation concentrations and tree diameter.

Total deposition

The total amount of ion deposition was calculated as kg ha^{-1} yr^{-1} using the precipitation and the concentrations for each sample. The annual SO_4^{-2} deposition in the open site plots was higher than the sum of the other anions at each elevation zone (*Fig. 4*). The amount of NO_2^-, F^-, and Br^- depositions in the open site plots remained below 1 kg across the elevation zones.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

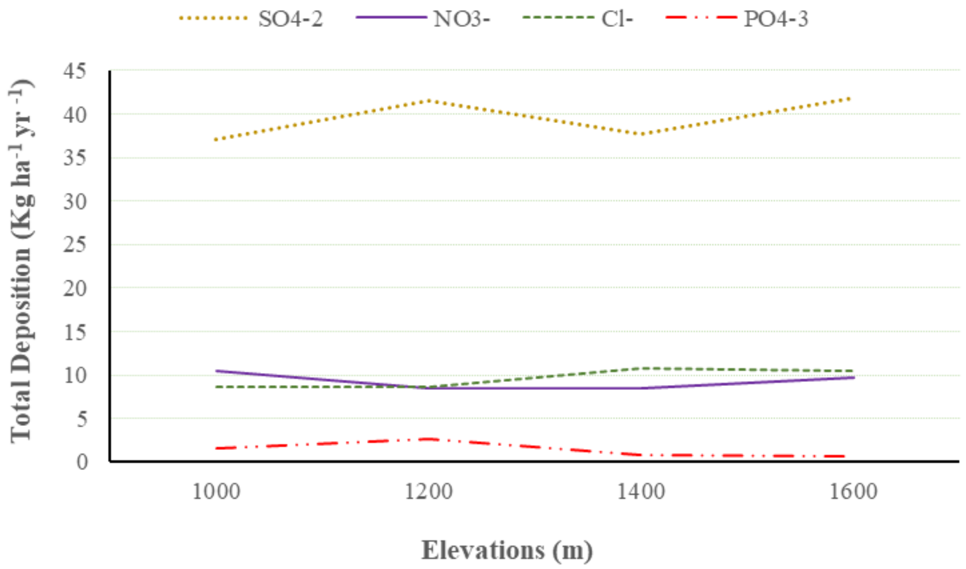

Figure 4. *Means ± standard deviation of open site total anion deposition (Kg ha^{-1} yr^{-1}) collected at different elevations (m) in Aladağ fir forest*

The open site Ca^{+2} and K^{+} depositions were higher than the other cations at each elevation zone. However, the open site NH_4^{+} deposition at 1200 m was about twice as high as that at the other elevation zones (*Fig. 5*).

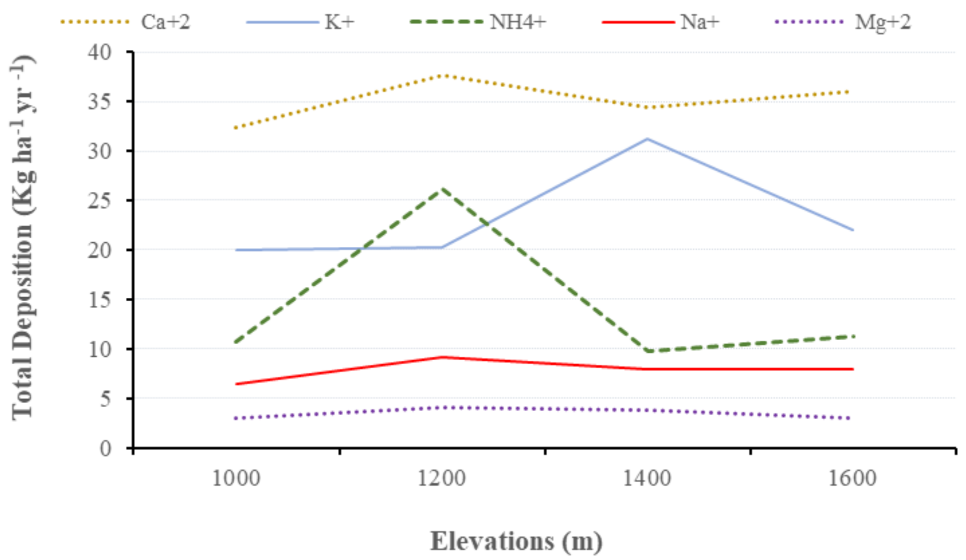

Figure 5. *Means ± standard deviation of open site total cation deposition (Kg ha^{-1} yr^{-1}) collected at different elevations (m) in Aladağ fir forest*

Throughfall SO_4^{-2} deposition was 50% higher at 1600 m compared to that at the other elevation zones. Throughfall NO_3^{-} deposition decreased by 75% above the 1000-m elevation zone (*Fig. 6*). Throughfall SO_4^{-2}, NO_3^{-}, Cl^{-}, and PO_4^{-3} deposition values were about 200%, 300%, 300%, and 400% higher than those of the adjacent open site plots at each elevation zone (*Fig. 6*).

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

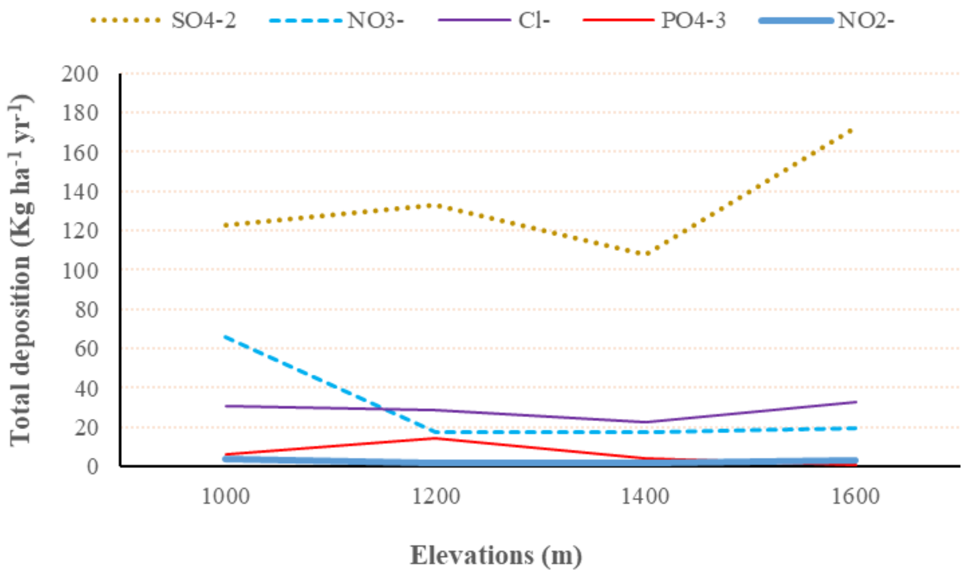

Figure 6. *Means ± standard deviation of throughfall total anion deposition (Kg ha^{-1} yr^{-1}) collected at different elevations (m) in Aladağ fir forest*

Throughfall K^+ deposition at 1200 m was about 40% higher than that at the other elevation zones (*Fig. 7*).

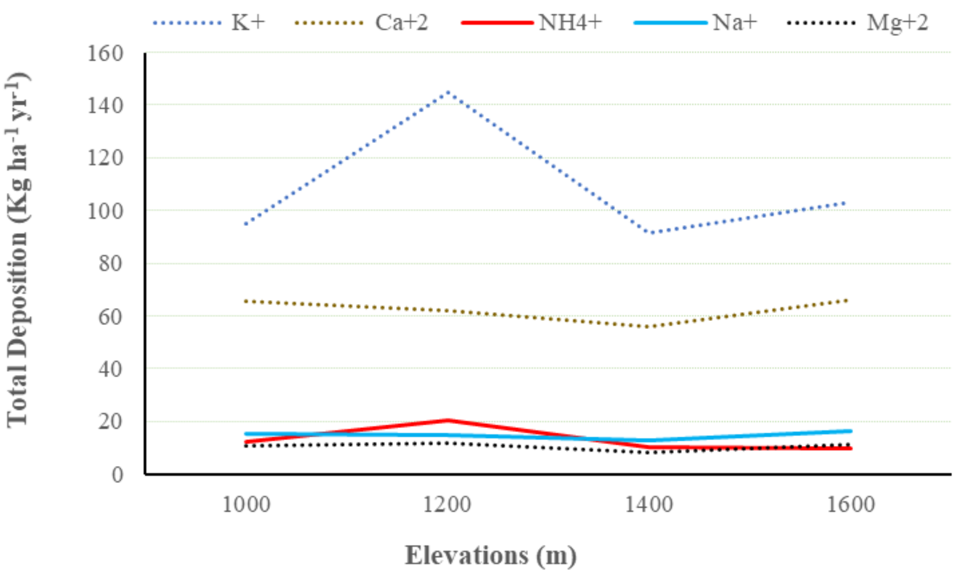

Figure 7. *Means ± standard deviation of throughfall total cation deposition (Kg ha^{-1} yr^{-1}) collected at different elevations (m) in Aladağ fir forest*

Throughfall K^+, Mg^{+2}, Na^+, Ca^{+2}, and NH_4^+ deposition values were about 400%, 300%, 200%, and 200% higher, respectively, than those of the adjacent open site plots at each elevation zone (*Fig. 7*).

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

Discussion

Canopy interception rate is related to the amount and intensity of precipitation, its distribution, and the stand type. In the current study, about 29% of the rainfall was intercepted by the fir canopy. In a study conducted by Garces et al. (2012) in the humid forests of Colombia having approximately 1500 mm of rainfall, the corresponding rate was about 19%. The differences between the current study and that of Garces et al. (2012) may be attributed to the stand type since the current study was carried out in a closed-canopy fir forest. Krupová et al. (2017) reported that the pH of precipitation collected between 2008 and 2010 was 5.5 and 6 in the spruce and fir forests of Slovakia, respectively. They reported a lower pH value in throughfall deposition compared to that of the open site plots. The higher pH value in the present study can be partially explained by the high base cation concentrations of the depositions.

In the current study, either the open site or the throughfall ion concentrations recorded at four elevation zones were within the specified ranges presented in the ICP-Forests Manual. However, the SO_4 and NO_3 anion and K and Ca cation values were higher compared to the other ions in the current study (*Table 3*).

Table 3. *Limits for some of the deposition variables in Part XIV of the ICP-Forests Manual and corresponding values in bulk depositions collected at different elevations (1000-1600 m) in Aladağ fir forest*

Variables	ICP-Min.	ICP-Max.	1000 m	1200 m	1400 m	1600 m
Reaction (pH)	2.5	9.4	6	6	6	6
EC (µS/cm)	1	10000	<100	<100	<100	<100
K (mg L^{-1})	0.002	250	8.2	11.8	7.07	7.2
Ca (mg L^{-1})	0.001	275	6.1	5.8	4.7	5.1
Mg (mg L^{-1})	0.0025	100	0.95	1	0.66	0.82
Na (mg L^{-1})	0.003	500	1.4	1.41	1.06	1.25
NH_4 (mg L^{-1})	0.002	175	1.3	2.3	0.96	0.89
Cl (mg L^{-1})	0.002	800	2.7	2.5	1.8	2.4
NO_3 (mg L^{-1})	0.002	175	5.5	1.55	1.36	1.52
SO_4 (mg L^{-1})	0.01	500	10.8	11.4	8.4	12.03
PO_4 (mg L^{-1})	0.0017	1000	0.49	1.21	0.31	0.07

Even though nitrogen is one of the most important plant nutrients in terrestrial ecosystems, until the 1980s its deposition was considered one of the main destructive agents in a majority of European forest. In addition to nitrogen, sulfate is also one of the most important air pollutants. Over-loaded NO_x and SO_x emissions from increased burning of fossil fuels and intensive agricultural and industrial activities had negatively affected European forest for several decades (Fisher et al., 2007; Erisman et al., 2008; Novotný et al., 2016; Thimonier et al., 2019). Following the framework agreement signed in 1979 to reduce the growing air pollution, atmospheric pollutants such as sulfur and nitrogen were targeted to remain within certain limits with the protocols of Aarhus 1998, Geneva 1984, Helsinki 1985, Soa 1988, and Gothenburg 1999. While some European countries have rapidly complied with the requirements of the agreement by preparing critical precipitation threshold maps, other countries have not been able to contribute sufficiently to reducing pollutants (Grennfelt et al., 2020). In Turkey, measures

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

concerning the release of industrial facility pollutants into the atmosphere were not as strictly regulated as in European countries. In addition, coal was used as fuel in most settlements in the Western Black Sea Region until 2005. Wood and coal are still used as the main fuel in rural areas. Although some air pollution variables are measured in terms of public health in city centers, there is insufficient data on the effects of these pollutants on forests, agriculture, and aquatic ecosystems, and the legal legislation to reduce them is either limited or inadequate. Therefore, pollutants released from settlements, intensive agricultural practices, and industrial activities can be transported by atmospheric movement and deposited on natural ecosystems in the vicinity of the source of the pollutants.

Research conducted in beech, spruce, and fir forests in central and western Europe has revealed that nitrogen deposition above 15-20 kg ha^{-1} may seriously affect above- and belowground ecosystem properties including biological diversity (Bobbink and Hettelingh, 2011; Schmitz et al., 2019). Waldner et al. (2015) stated that human-induced atmospheric sulfur and nitrogen deposition after the second half of the 20th century had acidified most of the forest soil. Schleppi et al. (2017) reported that 10-15 kg ha^{-1} annual nitrogen deposition in many European forests may accelerate nitrate leaching and result in water pollution. In the current study, the total open site and throughfall nitrogen deposition (NH_4 + NO_3) values were 200% and 300% more than those reported by Bobbink and Hettelingh (2011). Annual open site SO_4^{-2} deposition in the current study was about 40 kg/ha. Throughfall deposition was 300% higher than that of open site plots at 1000–1400-m elevation zones. The fate of the nitrogen deposited on forest soils depends on the interaction of many variables. The impact of nitrogen deposition on fir forests in the current study may not follow the same direction as many research findings in central and northern European forests. Excessive nitrogen deposition may lead to nutrient imbalance in saturated soils and it also increases plant sensitivity to insect and fungal diseases. Although there has been a problem of nitrogen saturation for many years in central and western European forests, each increment in nitrogen deposition has a negative impact on these forests (Schmitz et al., 2019; Etzold et al., 2020). On the other hand, poor soils may benefit from the fertilizing effect of nitrogen deposition (Waldner et al., 2015). Soil nutritional imbalance not only may impede the growth and physiological condition of plants, but it may also render forest ecosystems vulnerable to insect attacks. Yildiz et al. (2007a) reported that the fir-bark beetle (*Pityokteines curvidens* (Germ.)) and small fir-bark beetle (*Cryphalus piceae* (Ratz.)) cause a significant amount of tree loss in the Western Black Sea fir forests where the current study was conducted. They also reported that insect attacks decreased when the needle calcium content increased.

The acidifying effect of atmospheric depositions on soils varies depending on soil properties. For example, some soils with abundant base (K, Ca, and Mg) cations may display higher buffering capacity (Yildiz et al., 2007a). Yildiz et al. (2007b) reported that the soil in the region contains 400 kg ha^{-1} Ca, 160 ha^{-1} K, and 160 ha^{-1} Mg in the first 20-cm depth of the soil. The soil in the region has a substantial amount of acid neutralizing capacity for incoming atmospheric deposition. The anion and cation balance of the atmospheric deposition may also determine its acidifying capacity. By simply calculating the acid neutralization capacity (ANC) of the deposition at each sampling site using the *equations 1 and 2*:

$$(\mathrm{ANC}) = \sum (\text{base cations}) - \sum (\text{strong acid anions}) \qquad \text{(Eq.1)}$$

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

or

$$ANC = \sum (Ca + K + Mg + Na) - (SO_4 + NO_3 + Cl) \qquad \text{(Eq.2)}$$

The open site deposition ANC values at 1000-, 1200-, 1400-, and 1600-m elevation zones were found as 0.6, 1.31, 1.91, and 0.6, respectively. The ANC values of throughfall depositions were calculated as -3.36, 5.58, 1.95, and -2.41, respectively. Although the open site SO_4^{-2} deposition was high, base cations of the same depositions were also relatively high. Thus, the acid neutralization capacity of open site depositions remained high along all elevation zones. However, the acidifying potential of the throughfall depositions at 1000 and 1600 m was higher than at the other elevation zones.

Kimmins (1997) estimated that 45%, 35%, 66%, and 69% of the N, P, K, and Ca cycles, respectively, were biogeochemical cycles. Andesite, which constitutes the bedrock in the study sites, is mostly composed of plagioclase, hornblende, pyroxene, and biotite minerals. Plagioclase is one of the main sources of calcium in the soil. Augite and hornblende from pyroxenes also contain significant amounts of calcium. Biotite, also a component of andesite, contains a significant amount of potassium. Thus, a significant amount of calcium and potassium deposition may have local sources from the underlying bedrock. Kimmins (1997) also claimed that 31% of calcium and 12% of potassium have a geochemical cycle. Thus, some of the potassium and calcium deposited on the fir forests in the current study may have been transported from Central Anatolia via atmospheric movements. Stable isotope techniques can help to trace the source of the deposition. Demir et al. (2017) maintain that if the NO_3^- / SO_4^{-2} deposition ratio is less than “1”, then the pollutant source may be local. Since in the current study this ratio is about 0.24, settlements and recreational activities in the region may have been a source of the pollutants. Bayramoğlu Karşı et al. (2017) stated that coal and wood fuels are among the most important causes of atmospheric potassium deposition. Moreover, agricultural practices may be one of the important sources of nitrogen deposition near forest areas (Waldner et al., 2015; Kosonen et al., 2019). Thus, intensive poultry farming, recreational activities, and the use of wood and coal as a fuel in settlements around the current study sites may contribute to the atmospheric pollution in the region.

The cumulative effects of increased deposition over decades may make the forest ecosystem vulnerable to diseases and prone to further disturbances. Due to their biological structure, most lichen and algae species are sensitive to acid-forming pollutants such as sulfur and nitrogen. Upon conducting a research in a coniferous forest in Finland, Salemaa et al. (2020) reported that an annual nitrogen deposition of more than 5 kg ha^{-1} significantly affected algae species. Moreover, there has been a significant loss in lichen diversity in European forests due to air pollution (Agnan et al., 2017). Studies conducted by Agnan et al. (2017) in the forests of France and Switzerland revealed that the lichen diversity index is strongly related to eutrophication. In a study conducted by Şahin (2017) in the Aladağ fir forests of the current study, 56 lichen taxa belonging to 33 genera were identified. He found that the population of *Bryoria*, *Lobaria, Ramalina,* and *Usnea,* as species sensitive to air pollution, decreased at 900–1100-m elevation zones, where heavy sulfate and nitrogen depositions were reported in the current study. Şahin (2017) also claimed that pollutant-sensitive species such as *Evernia* and *Ramalina* had not been encountered around the recreational areas. Therefore, even though the annual deposition concentration falls within certain limits, the accumulation of nitrogen and sulfur components over many years may cause sensitive species to be adversely affected.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

The current study covered 1000–1600-m elevation zones. However, the ICP-Forests Level-II plots were established at 1600 m to monitor the regional fir forests. Therefore, the Level-II site located at only one elevation may not fully reflect the deposition on the fir forests of the region.

Conclusion and Suggestions

Sulfate, nitrogen, potassium, and calcium were the main ions deposited in the fir forests at the 1000–1600-m elevation zones in Aladağ. Even though only about 1/3 of the precipitation was intercepted, throughfall ion deposition was generally higher than the open site deposition, indicating that a significant amount of pollutant is captured by the canopy cover. This result implies the possibility of the filtering effects of forest canopy on pollutants deposited in openings around the forest. Therefore, instead of the forest openings, it would be more accurate to set up open site sampling plots above the canopy cover with the use of towers. The acid neutralization capacity of the deposition samples collected in the open site plots was high at all elevation zones. However, the acidifying capacity of the throughfall deposition at 1000 and 1600 m was higher than at the other elevation zones. Although the ion concentration values of the depositions remained within the ranges set by the ICP-Forests guidelines, the nitrogen and sulfur values of the current study were higher than those reported in many studies conducted in European forests. Different scenarios need to be developed addressing the problems that may arise from the long-term accumulation in the ecosystem, rather than focusing on annual concentration values. In addition, establishment of only one Level-II site at 1600 m for monitoring the condition might have been insufficient as a representation of fir forests. Therefore, either more Level-II monitoring plots should be established to represent different elevation zones or supporting studies should be conducted to increase the data validation.

Acknowledgements. This research was funded by the Duzce University BAP program through the project entitled “Bolu-Aladağ, Uludağ Göknarı (*Abies bormülleriana* Mattf.) Ormanlarında Atmosferik Çökelme [Atmospheric deposition in mature fir (*Abies bormülleriana* Mattf.) stands in Aladağ- Bolu]” and numbered as DÜBAP 02.02.213.

REFERENCES

[1] Agnan, Y., Probst, A., Séjalon Delmas, N. (2017): Evaluation of lichen species resistance to atmospheric metal pollution by coupling diversity and bioaccumulation approaches: A new bioindication scale for French forested areas. – Ecological Indicators 72: 99-110.

[2] Anonymous. (2010): ICP-Forests Manual: International Co-Operative Programme On Assessment and Monitoring of Air Pollution Effects on Forests, Part XIV, Sampling and Analysis of Deposition. – Institute for World Forestry, Hamburg.

[3] Anonymous. (2015): Türkiye Ormanlarının Sağlık Durumu (2008-2012). – Orman Genel Müdürlüğü, Ankara.

[4] Augustin, S., Bolte, A., Holzhausen, M., Wolff, B. (2005): Exceedance of critical loads of nitrogen and sulphur and its relation to forest conditions. – European Journal of Forest Research 124(4): 289-300.

[5] Bayramoğlu Karşı, M. B., Berberler, E., Karakaş, D. (2017): Gölcük Tabiat Parkı’ nda Yapılan Haftalık Atmosferik Toplam Çökelme Örneklerinin Polikistik Aromatik Hidrokarbon Anyon ve Katyon Konsantrasyonlarının Belirlenmesi ve Gölete Etkilerinin Araştırılması. – VII. Ulusal Hava Kirliliği ve Kontrolü Sempozyumu, 1-3 Kasım, Antalya.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

[6] Bobbink, R., Hettelingh, J. P. (2011): Review and Revision of Empirical Critical Loads and Dose-response Relationships. – Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM). www.rivm.nl/cce.
[7] Bussotti, F., Ferretti, M. (1998): Air pollution, forest condition and forest decline in Southern Europe: An overview. – Environmental Pollution 101: 49-65.
[8] Demir, T., Kılıçer, Y., Karakaş, S. (2017): Yarı Şehirsel İstasyonda Toplanan Yağmur Suyunun İyon Kompozisyonunun Belirlenmesi Ve Asitlik. – VII. Ulusal Hava Kirliliği ve Kontrolü Sempozyumu, 1-3 Kasım, Antalya.
[9] Erisman, J. W., Sutton, M. A., Galloway, J., Klimont, Z., Winiwarter, W. (2008): How a century of ammonia synthesis changed the World. – Nature Geoscience 1: 636-639.
[10] Etzold, S., Ferretti, M., Reinds, G. J., Solberg, S., Gessler, A., Waldner, P., Schaub, M., Simpson, D., Benham, S., Hansen, K., Ingerslev, M., Jonard, M., Karlsson, P. E., Lindroos, A. J., Marchetto, A., Manninger, M., Meesenburg, H., Merilä, P., Nöjd, P., Rautio, P., Sanders, T. G. M., Seidling, W., Skudnik, M., Thimonier, A., Verstraeten, A., Vesterdal, L., Vejpustkova, M., Vries, W. (2020): Nitrogen deposition is the most important environmental driver of growth of pure, even-aged and managed European forests. – Forest Ecology and Management 458: 117762.
[11] Fischer, R., Mues, V., Ulrich, E., Becher, G., Lorenz, M. (2007): Monitoring of Atmospheric Deposition in European Forests and an Overview on Its Implication on Forest Condition. – Applied Geochemistry 22: 1129-1139.
[12] Garces, B. M. L., Casas, F. A., Pena, M. (2012): Bulk Precipitation, Throughfall and Stemflow Deposition of N-NH_4, N-NH_3, and N-NO_3, In an Andean Forest. – Journal of Tropical Forest Secience 26(4): 446-457.
[13] Grennfelt, P., Engleryd, A., Forsius, M., Hov, Ø., Rodhe, H., Cowling, E. (2020): Acid rain and air pollution: 50 years of progress in environmental science and policy. – Ambio 49: 849-864.
[14] Kantarcı, M. D. (1980): Aladağ kütlesinin (Bolu) kuzey yamacındaki Uludağ Göknarı (*Abies bornmülleriana* Mattf.) Ekosistemlerinde Ekolojik Araştırmalar. – Orman Ekosistemi Sempozyumu, 10-15. Kasım, İstanbul.
[15] Kimmins, J. P. (1997): Forest Ecology: A Foundation for Sustainable Management (2nd Edition). – Prentice Hall, 1997.
[16] Kosonen, Z., Schnyder, E., Hiltbrunner, E., Thimonier, A., Schmitt, M., Seitler, E., Thöni, L. (2019): Current atmospheric nitrogen deposition still exceeds critical loads for sensitive, semi-natural ecosystems in Switzerland. – Atmospheric Environment 21: 214-225.
[17] Krupová, D., Bošeľa, M., Pavlenda, P., Tóthová, S. (2017): Long-term changes in atmospheric depositions in Slovakia. – Cent. Eur. For. J. 63(1): 42-47.
[18] Mayer, H., Aksoy, H. (1998): Türkiye Ormanları. – Muhtelif yayın 1. Baskı, Bolu, Türkiye, Batı Karadeniz Ormancılık Araştırma Enstitüsü Müdürlüğü Yayınları, Bolu.
[19] Novotný, R., Buriánek, V., Šrámek, V., Hůnová, I., Skořepová, I., Zapletal, M., Lomský, B. (2016): Nitrogen deposition and its impact on forest ecosystems in the Czech Republic - change in soil chemistry and ground vegetation. – iForest 10: 48-54.
[20] Salemaa, M., Kieloaho, A. J., Lindroos, A. J., Merilä, P., Poikolainen, J., Manninen, S. (2020): Forest mosses sensitively indicate nitrogen deposition in boreal background areas. – Environmental Pollution 261: 114054.
[21] SAS Systems for Windows™ (1996): Release 6.12. – SAS Institute Inc. Cary, North Carolina.
[22] Schleppi, P., Curtaz, F., Krause, K. (2017): Nitrate leaching from a sub-alpine coniferous forest subjected to experimentally increased N deposition for 20 years, and effects of tree girdling and felling. – Biogeochemistry 134: 319-33.
[23] Schmitz, A., Sanders, T. G., Bolte, A., Bussotti, F., Dirnböck, T., Johnson, J., Penuelas, J., Pollastrini, M., Prescher, A. K., Sardans, J., Verstraeten, A., Viries, W. D. (2019): Responses of forest ecosystems in Europe to decreasing nitrogen deposition. – Environmental Pollution 244: 980-994.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641

[24] Şahin, U. (2017): Aladağ göknar (*Abies nordmanniana* (Steven) Spach Subsp. *Equi-Trojani* (Asch. & Sint. Ex Boiss.) Coode & Cullen) Ormanlarında Epifitik Liken Çeşitliliği. – Düzce Üniversitesi Fen Bilimleri Enstitüsü, Orman Mühendisliği Anabilim Dalı. Yüksek Lisans Tezi.

[25] Thimonier, A., Kosonenb, Z., Braunc, S., Rihmd, B., Schleppia, P., Schmitta, M., Seitlerb, E., Waldnera, P., Thönib, L. (2019): Total deposition of nitrogen in Swissforests: Comparison of assessment methods and evaluation of changes over two decades. – Atmospheric Environment 198: 335-350.

[26] Waldner, P., Thimonier, A., Pannatier, E. G., Etzold, S., Schmitt, M., Marchetto, A., Rautio, P., Derome, K., Nieminen, T. M., Nevalainen, S., Lindroos, A., Merilä, P., Kindermann, G., Neumann, M., Cools, N., De Vos, B., Roskams, P., Verstraeten, A., Hansen, K., Karlsson, G. P., Dietrich, H. P., Raspe, S., Fischer, R., Lorenz, M., Iost, S., Granke, O., Sanders, T. G. M., Michel, A., Nagel, H. D., Scheuschner, T., Simoncic, P., Von Wilpert, K., Meesenburg, H., Fleck, S., Benham, S., Vanguelova, E., Clarke, N., Ingerslev, M., Vesterdal, L., Gundersen, P., Stupak, I., Jonard, M., Potocic, N., Minaya, M. (2015): Exceedance of critical loads and of critical limits impacts tree nutrition across Europe. – Annals of Forest Science 72(7): 929-939.

[27] Yıldız, O., Eşen, D., Akbulut, S. (2007a): Effects of different ecological and silvicultural factors on beetle catches in the Turkish fir (*Abies bornmulleriana* Mattf.) ecosystems. – J Pest Sci. 80: 145-150.

[28] Yıldız, O., Sarginci, M., Eşen, D., Cromack Jr., K. (2007b): Effects of vegetation control on nutrient removal and *Fagus orientalis*, Lipsky regeneration in the western Black Sea Region of Turkey. – Forest Ecology and Management 240: 186-194.

http://www.aloki.hu ● ISSN 1589 1623 (Print) ● ISSN1785 0037 (Online)
DOI: http://dx.doi.org/10.15666/aeer/1806_76277641