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Bulk precipitation chemistry at the forest and forest village

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ABSTRACT

The objectives of this study were to characterize and compare selected physical and chemical properties of precipitation in a broadleaf mixed forest and a village located in the same forest in order to determine the influence of the village on the atmospheric environmental quality of the forest ecosystem. Bulk precipitation samples were collected weekly from October 2005 to July 2011. Precipitation samples were analyzed for pH, electrical conductivity (EC), turbidity, total suspended particles (TSP), total alkalinity ($CaCO_3$), alkalinity (HCO_3^-), chloride (Cl^-), total hardness ($CaCO_3$), Ca hardness ($CaCO_3$), calcium (Ca^{2+}), magnesium (Mg^{2+}), organic matter, total nitrogen (N), sodium (Na^+), potassium (K^+), nitrate $(NO_3)^-$, phosphate $(PO_4)^{3-}$, iron (Fe), aluminum (Al), ammonium $(NH_4)^+$, and sulfate $(SO_4)^{2-}$. The precipitation samples from forest and forest village were significantly different from each other for EC, total alkalinity ($CaCO_3$), HCO_3^-, Ca hardness ($CaCO_3$), Ca^{2+}, total N, K^+, NO_3^-, and NH_4^+. Regardless of the study sites, PO_4^{3-} and Mg^{2+} were dominant anion and cation, respectively. The orders of cation and anion concentrations were similar for both study sites and they were in descending order of $Mg^{2+} > Ca^{2+} > Na^+ > K^+ > NH_4^+ > Fe > Al > H^+$ and $PO_4^{3-} > HCO_3^- > SO_4^{2-} > Cl^- > NO_3^-$, respectively. Overall average monthly values of precipitation characteristics did not show significant difference between months except for pH, EC, total alkalinity ($CaCO_3$), and HCO_3^-. Significant correlations were found between SO_4^{2-} and pH, Ca^{2+}, NH_4^+ and between NO_3^- and NH_4^+ for the precipitation event at the forest site. Both study sites had pH values higher than 5.6 due to the neutralization of SO_4^{2-} and NO_3^- by NH_4^+ and Ca^{2+}. Significant correlation coefficients found between the study sites for the same precipitation parameter indicated that both study sites were under the influence of the same emission sources.

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1. Introduction

Population growth and industrial developments cause increases in the concentrations of pollutants in the atmosphere. These pollutants return to the ground through precipitation in two forms as dry and wet depositions. Both deposition forms together are known as bulk deposition (Akkoyunlu and Tayanç, 2003; Herrera et al., 2009). Therefore, precipitation chemistry has received more attention among scientists and many studies have been conducted around the world (Alastuey et al., 1999; Gülsoy et al., 1999; Fujita et al., 2000; Miller et al., 2000; Lara et al., 2001; Driscoll et al., 2003; Hu et al., 2003; Zhang et al., 2003; Zunckel et al., 2003; Kang et al., 2004; Das et al., 2005; Mouli et al., 2005; Staelens et al., 2005; Sakihama et al., 2008; Song and Gao, 2009; Yi et al., 2010; Cheng et al., 2011). Concentrations and dominance of the ions varied in these studies and the results were not consistent with each other due to the differences in the characteristics of study sites (urban vs. rural area), intensity of industrial facilities, sampling types (wet-only vs. bulk deposition), sampling seasons and intervals, locations of the study sites in relation to main air masses, and topographic and climatic conditions (Heuer et al., 2000; Tuncer et al., 2001; Akkoyunlu and Tayanç, 2003; Staelens et al., 2005; Tu et al., 2005; Luo et al., 2007; Yi et al., 2010). In the majority of

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these studies, precipitation was typically acidic with pH values lower than 5.6 (Fujita et al., 2000; Lara et al., 2001; Akkoyunlu and Tayanç, 2003; Hu et al., 2003; Migliavacca et al., 2004; Staelens et al., 2005; Luo et al., 2007; Sakihama et al., 2008; Yi et al., 2010). Alkaline character with pH values greater than 5.6 was observed in a limited number of studies (Alastuey et al., 1999; Tuncer et al., 2001; Topçu et al., 2002; Bayraktar and Turalioglu, 2005; Mouli et al., 2005; Das et al., 2005). Also, precipitation alkalinity in these studies was attributed to the neutralization of acidity by cations, ammonia and calcium salts, and carbonate rich air masses influencing study sites (Alastuey et al., 1999; Gülsoy et al., 1999; Topçu et al., 2002; Akkoyunlu and Tayanç, 2003; Safai et al., 2004; Bayraktar and Turalioglu, 2005; Mouli et al., 2005; Sakihama et al., 2008). Acidity of the rainfall is associated with sulfate and nitrate emissions (Das et al., 2005; Sakihama et al., 2008) and acid rains cause decreases in the diversity and abundance of floral and faunal species in the soil and aquatic environments (Heuer et al., 2000; Lara et al., 2001; Driscoll et al., 2003; Sakihama et al., 2008).

Besides long range gas and aerosol emissions, the chemical composition of bulk deposition can also be affected by local gas and aerosol emissions which are influenced by degree of industrialization, climatic conditions, weather and geographic conditions in a specific site (Staelens et al., 2005; Sakihama et al., 2008). In fact, Alastuey et al. (1999) found significant correlations between concentrations of SO_4^{2-} and NH_4^+ in the precipitation and local sources of SO_4^- and NH_4^+. On the other hand, wet deposition can be composed of regional emissions depending on the movements of air masses (Kubilay et al., 2000). For instance, alkaline pH value of the precipitation in the Mediterranean area was attributed to carbonate rich air mass intrusions from Africa (Quereda et al., 1996). Concentrations of the precipitation parameters in bulk deposition can be higher than the concentrations of the same parameters in wet deposition since bulk deposition consists of both dry and wet depositions, and soil originated particles can be deposited on the walls of the samplers in dry periods when bulk deposition samplers are exposed to the atmosphere for a long period of time (Akkoyunlu and Tayanç, 2003; Bayraktar and Turalioglu, 2005; Staelens et al., 2005). On the other hand, ions in the precipitation can originate from different sources such as seawater, anthropogenic and crustal sources. For instance, Okay et al. (2002) found that the majority of Ca^{2+} ions in the precipitation came from the soil, while Mg^{2+} and K^+ ions were derived from the seawater. In a similar study, Sakihama et al. (2008) suggested that Na^+, K^+, Ca^{2+}, Mg^{2+}, and Cl^- ions in the precipitation at Okinawa Island of Japan originated from sea salts due to the typhoons and wind-blown soil particles. Cheng et al. (2011) determined that Na^+ and Cl^- ions were mainly derived from seawater; Mg^{2+} from seawater and crustal sources, and NH_4^+, K^+, Ca^{2+}, NO_3^-, and SO_4^{2+} from the anthropogenic sources.

Villages located in the forest ecosystems release pollutants into the atmosphere which influence the air quality and the health of forest ecosystems. Chemical contributions from these types of residential areas can reach alarming levels depending on the characteristics of emission sources. Several bulk and wet deposition studies were carried out at various regions of Turkey, especially in urban sites and they did not include forest areas or compare atmospheric chemistry of forestland with those of residential areas located in the forest ecosystems (Gülsoy et al., 1999; Kubilay et al., 2000; Okay et al., 2002; Akkoyunlu and Tayanç, 2003; Başak and Alagha, 2004). Studies from different parts of the world on precipitation chemistry were also conducted mostly in residential areas or a mixture of various land use forms other than rural areas or forestlands. Some studies on precipitation chemistry were also carried out in rural sites located around industrial facilities (Alastuey et al., 1999; Migliavacca et al., 2004). However, compared to urban studies, there is limited number of studies implemented in rural areas. For instance, Heuer et al. (2000) studied precipitation chemistry in the central Rocky Mountains and found that precipitations on western slope of the Rocky Mountains had greater concentrations of NO_3^- and SO_4^{2-} in winter while the precipitations on the eastern slopes had greater concentrations of NO_3^- and NH_4^+ in summer. Cheng et al.(2011) also studied precipitation chemistry in a mountainous region in China and determined that precipitation was acidic, and sources of NH_4^+ K^+, Ca^{2+}, NO_3^-, and SO_4^{2-} were related to anthropogenic activities. In general, the main purposes of these studies were to determine the chemical composition of the precipitation and to identify possible sources of the major ions in the rainwater (Lara et al., 2001; Safai et al., 2004; Mouli et al., 2005; Tu et al., 2005; Herrera et al., 2009; Song and Gao, 2009). Therefore, the objectives of our study were to characterize the chemical composition of bulk precipitation events at both forest and forest village sites, and to compare precipitation chemistry of the forest site with that of the forest village, Bahçeköy, to find out the influence of emissions from the forest village on atmospheric environmental quality of the forest ecosystem.

2. Material and methods

2.1. Study site

This study was conducted in a dense old growth mixed broadleaf deciduous forest ecosystem in Belgrad Forest and in a forest village named Bahçeköy located in the same forest ecosystem. The distance between study sites in the forest and forest village was about 8.3 km. Bahçeköy is one of the most populated forest villages in Turkey with a population of about 13 thousand and approximately 5 thousand households. There is no industrial facility and agricultural activity in the forest village. Exhaust gasses from vehicles and fossil fuels used for heating and cooking are the main sources of gasses and aerosol emissions into the atmosphere in the region. Forest vegetation in the study site is composed of woody species such as *Quercus frainetto* Ten., *Quercus cerris* L., and *Fagus orientalis* L. mixed with varying amounts of *Populous tremula* L., *Ulmus campestris* L., *Alnus glutinosa* L., *Acer trautvetteri* Med., *Carpinus betulus* L., *Castanea sativa* Mill., *Acer campestre* L., and *Sorbus torminalis* Crantz. (Yaltırık, 1966). The study site has a soil type of Vertic Xerochrept developed from mainly Neocene deposits and carboniferous clay schists (USDA, 1996). The schists underlying the Neocene deposits are rich in glauconite containing FeO, Fe_2O_3, Al_2O_3, MgO, CaO, and K_2O (Özhan, 1977).

According to the Thornthwaite classification method, the study site has a humid, mesothermal and maritime climate with a moderate water deficit in summer (Özyuvacı, 1999). Average annual precipitation is about 1129 mm and potential

evapotranspiration varies between 752 and 833 mm (Özhan et al., 2010). Average annual temperature is about 12.3 °C and the warmest month is August with an average temperature of 21.7 °C, while the coldest month is February with an average temperature of 4.2 °C in the study area (Özhan et al., 2008). Prevailing NW maritime winds influence study sites, especially in winter (Balcı et al., 1986).

2.2. Sampling and analysis

Bulk precipitation samplers were installed at two permanent forest openings with sizes of 100 m × 50 m in the Belgrad Forest (41°13′ 00″–41°14′ 13″N, 28°54′ 25″–28°56′ 37″E) and on the roof of a 5 m-high building in the Faculty of Forestry campus of Istanbul University in Bahçeköy (41°09′ 57″–41°11′ 01″ N 28°59′ 10″–28°59′ 57″E). Three rainwater collectors consisting of polyethylene collection bottles (3.5 L in volume) and funnels (22.6 cm in diameter) attached to the stakes were placed 1.5 m above the ground at the forest sites and the roof of the building for each study site. Bulk precipitation samples were collected on a weekly basis from October 2005 to July 2011. The funnels were covered with polyethylene net sieves (with 1 mm mesh width) and the collection bottles were placed in a PVC pipe (15 cm in diameter) at the study sites in order to prevent contamination and keep samples in the dark for a one week period.

During the sample collection, the contents of three bottles were mixed and one 1.5- or 2-liter subsample was obtained for each study site depending on the presence and amount of the precipitation. At the laboratory the samples were filtered through a filter paper. Then 1-liter samples were separated and stored in pre-cleaned polyethylene bottles in the refrigerator at 4 °C until conducting chemical analysis (Akkoyunlu and Tayanç, 2003; Bayraktar and Turalioglu, 2005) and the rest of the water sample was analyzed for pH, EC, turbidity, organic matter, TSP, total hardness, Ca hardness, HCO_3^-, and Cl^-. Stored rainwater samples were analyzed for total alkalinity ($CaCO_3$), Ca^{2+}, Mg^{2+}, total N, Na^+, K^+, NO_3^-, PO_4^{3-}, Fe, Al, NH_4^+, and SO_4^{2-}.

The chemical analyses were usually conducted within one month after sample collection. After removing the samples from the collection bottles in the laboratory, the sampling equipment was cleaned with distilled water and kept clean until the following sampling week. When the amount of precipitation was insufficient for the analysis, polyethylene collection bottles and funnels were replaced with clean collectors.

Laboratory analyses were conducted according to the standard methods of the American Public Health Association (APHA, 1998). The pH and EC values were determined by methods 4500-HB and 2510 B, respectively using the WTW Multiline P4 Universal Meter (WTW, Weilheim, Germany). Turbidity was measured by Method 2130 B using Orbecohellige 965-IR turbidity meter and TSP was determined according to the evaporation procedure performed on 100 mL rainwater samples with an accuracy of 0.1 mg. Alkalinity and hardness were analyzed according to 2320 B titration method and 2340 C EDTA titrimetric method, respectively. Ca^{2+}, Mg^{2+}, and Cl^- were analyzed with 3500-Ca BEDTA titrimetric, 3500-Mg B calculation and 4500-C1-B argentometric methods, respectively. Total N was determined according to 4500-Norg C semi-micro-kjeldahl method using a Velp UDK 132 distillation unit. The concentration of NO_3^- was measured by 4500-NO_3 D nitrate electrode method and NH_4^+ by 4500-NH_3 D ammonium selective electrode method using Thermo-orion 720 A ion meter. SO_4^{2-} was analyzed by 4500-SO_4 E turbidimetric method, PO_4^{3-} by 4500-P C vanadomolybdophosphoric acid colorimetric method, Al by 3500-A1 B eriochrome cyanine R method, and Fe by 3500-Fe B phenanthroline method using Shimadzu UV-1700 spectrophotometer. Na^+ was analyzed according to AOAC official method 976.25 (AOAC International, 1990) and K^+ according to 3500 K C Potassium-selective electrode method using Thermo-orion 720A ion meter. Additionally, pH values were used to calculate the concentration of H^+ ions (Zhang et al., 2003; Migliavacca et al., 2004; Cheng et al., 2011). Finally, organic matter was analyzed with respect to TS 6288 EN ISO 8467 (Turkish Standard Institute, 1998).

To analyze the differences between the study sites, among the months, and variations of the precipitation characteristics for each study site with respect to the months, data were analyzed using two-way ANOVA test and sample means were compared with Tukey test. Prior to ANOVA, data were tested for normality and logarithmic transformation was performed on data if necessary (Zar, 1996). Pearson correlation analysis was also performed on the data to determine possible relationships between the rainwater parameters for each study site separately and between the forest and forest village study sites for the same precipitation characteristic (Zar, 1996).

3. Results

Concentrations of major chemical components, along with pH, turbidity, total TSP, and organic matter for bulk samples were given in Table 1 for the forest and forest village study sites. The forest and forest village study sites differed significantly for EC, total alkalinity ($CaCO_3$), HCO_3^-, Ca hardness ($CaCO_3$), Ca^{2+}, total N, K^+, NO_3^-, and NH_4^+ ($P < 0.05$). Precipitation at the forest village had significantly lower EC, total alkalinity ($CaCO_3$), HCO_3^-, K^+, and NH_4^+ values than the precipitation at the forest site. The orders of cation and anion concentrations were similar for both study sites and they were in descending order of $Mg^{2+} > Ca^{2+} > Na^+ > K^+ > NH_4^+ > Fe > Al > H^+$ and $PO_4^{3-} > HCO_3^- > SO_4^{2-} > CI^- > NO_3^-$, respectively (Table 1). Regardless of the study sites, PO_4^{3-} was the dominant anion with average concentrations of 26.98 mg/L for the forest and 23.13 mg/L for the forest village study sites. Mg^{2+} was the dominant cation with average concentrations of 7.15 mg/L for the forest and 7.98 mg/L for the forest village study sites (Table 1). In contrast, H^+ had the lowest concentration in the precipitation at both study sites. Overall, average monthly values of rainwater quality parameters averaged over study sites did not show significant differences between months except for the pH (Fig. 1a), H^+ (Fig. 1b), EC (Fig. 1c), Fe (Fig. 1d), NH_4^+ (Fig. 1e), total alkalinity ($CaCO_3$), and HCO_3^-.

On the other hand, the results revealed that in contrast to the effects of main factors, the intra-site differences of the ionic concentrations of precipitation were not statistically significant except for total alkalinity ($CaCO_3$) and HCO_3^- (Fig. 2). All rainwater parameters showed an almost similar trend in both study sites throughout the year except for the total alkalinity ($CaCO_3$) and HCO_3^- and annual trend of some

Table 1
Chemical composition of bulk precipitation in the study sites (mean ± SEM).

Rainwater quality parameters	Study sites		Probability
	Forest	Forest village	
pH	6.47 ± 0.09	6.67 ± 0.11	P > 0.05
H^+ (μg/L)	2.00 ± 1.00	1.80 ± 1.00	P > 0.05
EC (μS/cm)	81.33[a*] ± 7.21	76.04[b] ± 9.95	P < 0.001
Turbidity (NTU)	8.34 ± 1.40	16.74 ± 4.57	P > 0.05
TSP (mg/L)	0.04 ± 0.005	0.04 ± 0.005	P > 0.05
Total alkalinity ($CaCO_3$) (mg/L)	12.88[a] ± 1.66	9.94[b] ± 1.23	P < 0.001
HCO_3^- (mg/L)	18.48[a] ± 2.48	12.12[b] ± 1.50	P < 0.001
Cl^- (mg L^{-1})	6.30 ± 0.65	4.80 ± 0.48	P > 0.05
Total hardness ($CaCO_3$) (mg/L)	42.18 ± 2.19	46.87 ± 3.63	P > 0.05
Ca Hardness ($CaCO_3$) (mg/L)	13.08[a] ± 1.18	14.11[b] ± 1.75	P < 0.001
Ca^{2+} (mg/L)	5.27[a] ± 0.47	5.75[b] ± 0.70	P < 0.001
Mg^{2+} (mg/L)	7.15 ± 0.42	7.98 ± 0.72	P > 0.05
Organic matter (mg/L)	3.74 ± 0.49	2.68 ± 0.26	P > 0.05
Total N (mg/L)	0.58[a] ± 0.06	0.65[b] ± 0.08	P < 0.001
Na^+ (mg/L)	2.26 ± 1.19	3.59 ± 2.27	P > 0.05
K^+ (mg/L)	1.00[a] ± 0.16	0.98[b] ± 0.12	P < 0.001
NO_3^- (mg/L)	1.88[a] ± 0.29	2.17[b] ± 0.33	P < 0.001
PO_4^{3-} (mg/L)	26.98 ± 2.42	23.13 ± 2.55	P > 0.05
Fe (μg/L)	222.73 ± 29.11	245.63 ± 54.65	P > 0.05
Al (μg/L)	96.48 ± 18.31	61.05 ± 18.54	P > 0.05
NH_4^+ (mg/L)	0.39[a] ± 0.05	0.37[b] ± 0.03	P < 0.001
SO_4^{2-} (mg/L)	7.69 ± 1.06	6.78 ± 0.96	P > 0.05

*Means with different superscript letters are significantly different at alpha level of 0.05 as determined by Tukey test (P < 0.05).

ions for forest and forest village study sites were presented in Fig. 3. Therefore, we focused on the effects of main factors and significant interactions instead of presenting results of insignificant interactions between study sites and months.

Rainwater was alkaline for both study sites with average pH values of 6.47 and 6.67 for the forest and forest village, respectively, and they were not significantly different from each other (Table 1). The average monthly pH values showed significant differences and ranged from 5.95 in November to 7.15 in April (Fig. 1a). Rainwater pH showed significant correlations with half of the parameters studied including SO_4^{2-} (r = 0.46, P < 0.01), organic matter (r = 0.44, P < 0.01), and Ca^{2+} (r = 0.57, P < 0.01) for the forest site (Table 3) while it did not show significant correlations with more than half of the parameters including SO_4^{2-} and organic matter for the forest village study site (Table 4). There was no significant difference between study sites for H^+ concentration, which was 2.00 μg/L and 1.80 μg/L for the forest and the forest village sites, respectively (Table 1). Concentration of H^+ differed significantly with respect to months with the highest value in February and the lowest value in April (P < 0.05; Figs. 1b and 3b). Compared to the other months, higher H^+ concentrations in winter months explain the lower pH values in the corresponding months (Fig. 3a and b).

Average EC values were 81.33 μS/cm and 76.04 μS/cm for the forest and the forest village study sites, respectively, and these values were significantly different from each other (Table 1; P < 0.001). Overall mean monthly EC values averaging over study sites showed significant differences among the months and ranged from 43.85 μS/cm in November to 133.99 μS/cm in August (Fig. 1c; P < 0.002). Also, significant correlations existed between EC and most of the rainwater quality parameters including pH, turbidity, TSP, Cl^-, Ca^{2+}, organic matter, and SO_4^{2-} for the precipitation at the forest site and pH, TSP, Cl^-, Ca^{2+}, organic matter, Fe, Al, NH_4^+, and SO_4^{2-} for the precipitation at the forest village site (Tables 3 and 4).

The turbidity, TSP, and organic matter values of the precipitation did not show significant differences with respect to the study sites and to the months. Turbidity values were 8.34 Nephelometric Turbidity Unit (NTU) for the forest site and 16.74 NTU for the forest village site (Table 1). The overall average monthly turbidity values averaging over study sites ranged between 4.57 and 24.51 NTU (Table 2). Turbidity value showed significant correlations with TSP (r = 0.49, P < 0.01), total alkalinity ($CaCO_3$) (r = 0.39, P < 0.01), HCO_3^- (r = 0.47, P < 0.01), Ca hardness (r = 0.50, P < 0.01), Ca^{2+} (r = 0.51, P < 0.01), organic matter (r = 0.48, P < 0.01), Fe (r = 0.52, P < 0.01), and SO_4^{2-} (r = 0.36, P < 0.05) for the precipitation at the forest site (Table 3) while it did not show significant correlation with any rainwater quality parameter for the precipitation at the forest village site (Table 4).

Concentration of TSP in the precipitation was low and both study sites had similar TSP values (0.04 mg/L) (Table 1). Overall mean monthly TSP values were also very low and ranged between 0.02 and 0.06 mg/L (Table 2). Concentration of TSP had significant correlations with 14 out of 21 parameters including pH (r = 0.32, P < 0.05), Ca^{2+} (r = 0.71, P < 0.01), organic matter (r = 0.58, P < 0.01), and SO_4^{2-} (r = 0.75, P < 0.01) for the precipitation at the forest site (Table 3) and with 7 out of 21 parameters including organic matter (r = 0.58, P < 0.01), Fe (r = 0.56, P < 0.01), and SO_4^{2-} (r − 0.54, P < 0.01) for the precipitation at the forest village study site (Table 4).

The concentration of the organic matter in the precipitation was also low and similar for the forest and forest village study sites (Table 1). Overall average monthly concentrations of the organic matter varied between 2.05 and 7.52 mg/L (Table 2). The concentration of organic matter had significant correlations with most of the rainwater parameters but it had relatively high correlations with Cl^- (r = 0.73, P < 0.01), Ca hardness (r = 0.78, P < 0.0.01), Ca^{2+} (r = 0.78, P < 0.01) and SO_4^{2-} (r = 0.67, P < 0.01) for the precipitation at the forest site possibly indicating that they may come from the same source (Table 3). In contrast to the forest site, the concentration of organic matter showed significant correlations with less rainwater parameters for the precipitation at the forest village site (Table 4). Total alkalinity ($CaCO_3$) showed significant variations with respect to the study sites and months (P < 0.05; Table 1). The forest study site had a higher concentration of total alkalinity in the precipitation than the forest village site (Table 1), and overall average monthly concentration of total alkalinity ranged between 5.80 and 23.01 mg/L. Also, interaction between the main factors was significant for the concentration of the total alkalinity ($CaCO_3$) in the precipitation (P < 0.05). The concentrations of the total alkalinity ($CaCO_3$) in the precipitation at both study sites followed also similar trend, and the forest study site had a significantly higher mean total alkalinity ($CaCO_3$) than the forest village during the winter months (Fig. 2a). HCO_3^- also showed similar trend to the total alkalinity ($CaCO_3$) and the forest site had a higher HCO_3^- content than the forest village site in the precipitation (Table 1). Significant interaction was also found

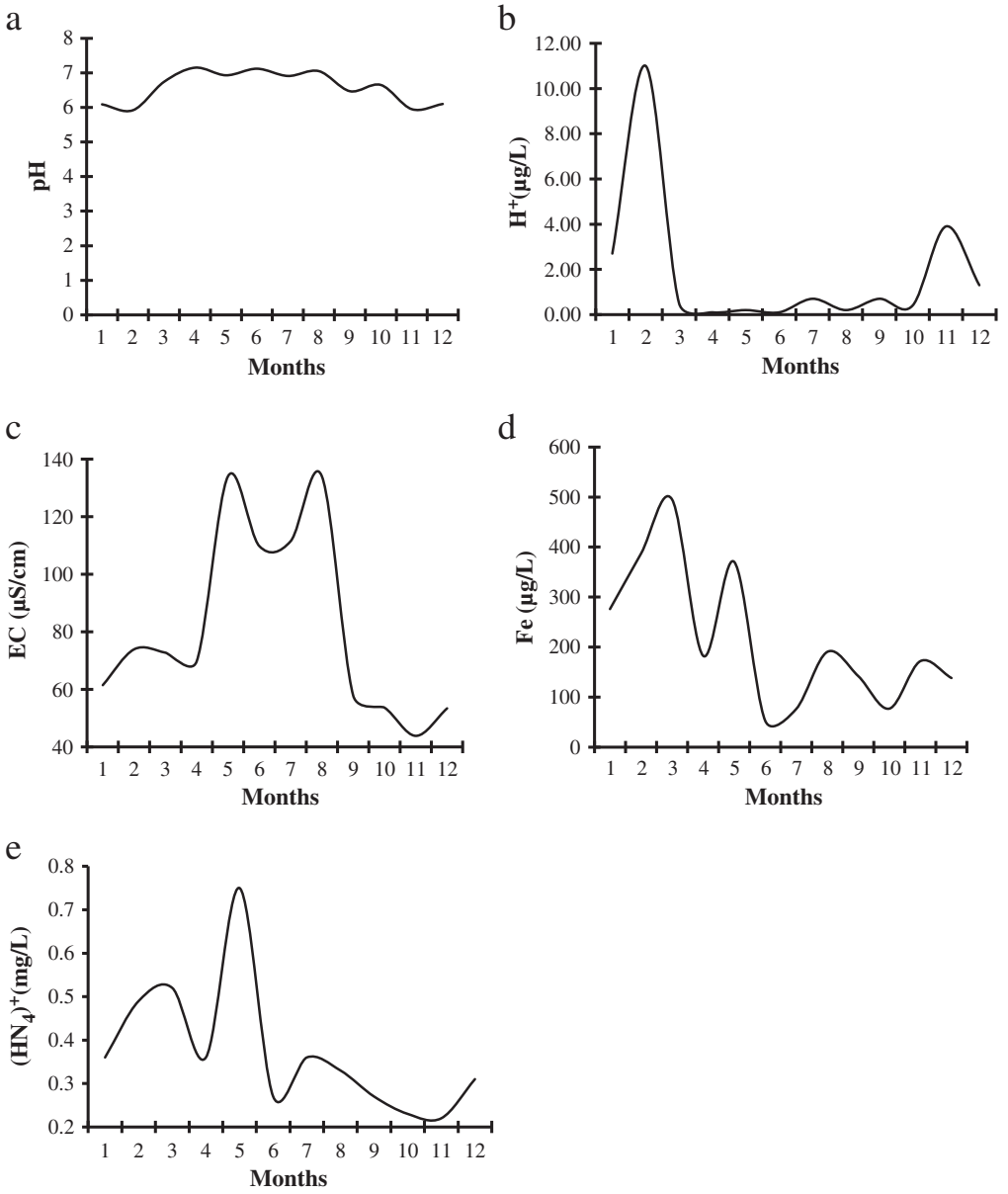


Fig. 1. Overall average monthly pH, H^+, EC, Fe, and NH_4^+ values with respect to months.

between study sites and months for the concentration of HCO_3^- in the precipitation ($P < 0.05$). Although HCO_3^- content of the rainwater showed a similar trend at both study sites (Fig. 2b) it was significantly higher in the precipitation at the forest site than at the forest village during the summer and winter months. Both total alkalinity and HCO_3^- had significant correlations with most of the precipitation parameters including SO_4^{2-} for both forest and forest village sites (Tables 3 and 4). Moreover, HCO_3^- showed good correlations with both K^+ and Ca^{2+} for the precipitation at the forest site (Table 3) and only with Ca^{2+} for the precipitation at the forest village (Table 4).

The concentration of Cl^- ion did not differ between study sites and it was 6.30 mg/L for the forest and 4.80 mg/L for the forest village (Table 1). The concentration of Cl^- did not show significant change and remained similar throughout the year (Table 2). Similar to Cl^-, the concentration of Na^+ also did not show significant differences with respect to the study sites (Table 1) and months (Table 2), and monthly average values varied between 0.65 and 14.14 mg/L. Although it is assumed that the Na^+ and Cl^- ions originated from the sea salts, significant correlations were not found between Na^+ and Cl^- ions for the precipitations either at the forest or the forest village sites (Tables 3 and 4). Additionally, Na^+ did not have significant correlations with any other rainwater quality parameters except for NO_3^- for the forest study site (Table 3) and K^+ for the forest village study site (Table 4). Contrary to Na^+ ion, Cl^- ion showed significant correlations with most of the parameters including Ca^{2+} ($r = 0.68$, $P < 0.01$), organic

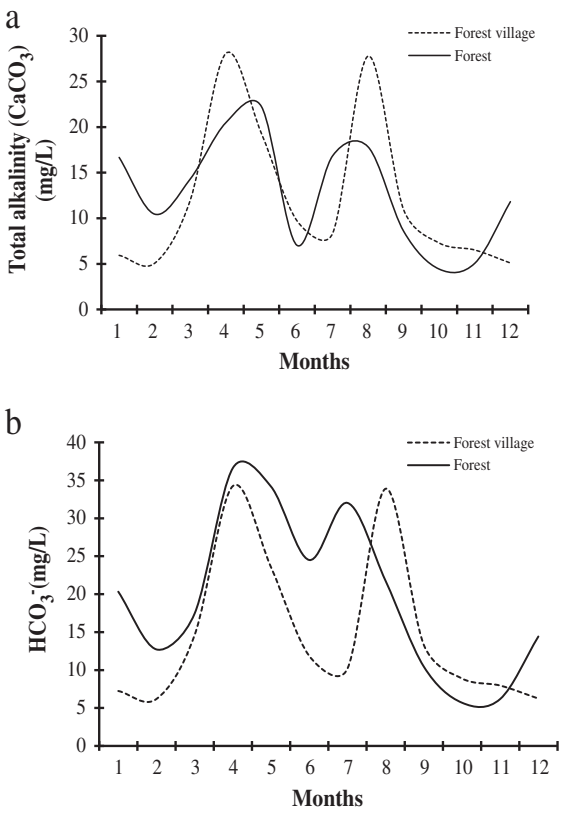


Fig. 2. Average monthly concentration of total alkalinity ($CaCO_3$) and HCO_3^- in the precipitation at the forest and forest village study sites.

matter ($r = 0.73$, $P < 0.01$), K^+ ($r = 0.54$, $P < 0.01$), PO_4^{3-} ($r = 0.59$, $P < 0.01$), and SO_4^{2-} ($r = 0.54$, $P < 0.01$) for the precipitation at the forest site (Table 3) and with Ca^{2+} ($r = 0.54$, $P < 0.01$), organic matter ($r = 0.50$, $r = 0.01$), K^+ ($r = 0.36$, $P < 0.05$), and SO_4^{2-} ($r = 0.61$, $P < 0.01$) for the precipitation at the forest village study site (Table 4).

Among the base cations, the concentrations of Ca^{2+} and K^+ in the precipitation significantly differed while Mg^{2+}did not with respect to the study sites. The forest site had a significantly higher K^+ but lower Ca^{2+} values than the forest village whereas both sites had similar amount of Mg^{2+} concentration in the rainwater (Table 1). The overall average monthly concentrations of all three base cations did not show significant variation throughout the year and ranged between 2.67 and 8.90 mg/L, 5.65 and 10.74 mg/L, and 0.20 and 1.91 mg/L for Ca^{2+}, Mg^{2+}, and K^+, respectively (Table 2). The concentration of Ca^{2+} ion showed high correlations with concentrations of SO_4^{2-} ($r = 0.74$, $P < 0.01$), organic matter ($r = 0.78$, $P < 0.01$), and TSP ($r = 0.71$, $P < 0.01$) for the rainwater at the forest site (Table 3) and organic matter ($r = 0.42$, $P < 0.01$) and SO_4^{2-} ($r = 0.42$, $P < 0.01$) for the rainwater at the forest village site (Table 4). Compared to Ca^{2+}, Mg^{2+} ion had significant correlations with less rainwater parameters for both forest (Table 3) and forest village sites (Table 4). Similar to Mg^{2+}, K^+ also showed significant correlations with a few rainwater parameters including TSP, HCO_3^-, Cl^-, Ca hardness, Ca^{2+}, organic matter, NH_4^+, and SO_4^{2-} in the forest site (Table 3) and Cl^- and Na^+ in the forest village (Table 4).

Concentrations of the organic matter in the precipitations were similar for both study sites, and they were 3.74 mg/L and 2.68 mg/L for the forest and forest village sites, respectively (Table 1). On the other hand, overall average monthly organic matter contents of the rainwater averaging over study sites ranged from 2.05 mg/L to 7.52 mg/L and they did not show significant differences (Table 2). Significant correlations existed between organic matter content and some ions including Cl^- ($r = 0.73$, $P < 0.01$), Ca^{2+} ($r = 0.78$, $P < 0.01$), K^+ ($r = 0.62$, $P < 0.01$), PO_4^{3-} ($r = 0.54$, $P < 0.01$), and SO_4^{2-} ($r = 0.67$, $P < 0.01$) for the precipitation at the forest site (Table 3) and Cl^- ($r = 0.50$, $P < 0.01$), Ca^{2+} ($r = 0.42$, $P < 0.01$), Fe ($r = 0.36$, $P < 0.05$), and SO_4^{2-} ($r = 0.55$, $P < 0.01$) for the precipitation at the forest village (Table 4).

There was a significant difference between total N concentration of the precipitation at the forest and forest village sites ($P < 0.05$) and the precipitation at the forest village had a significantly higher total N concentration (0.65 mg/L) than that at the forest site (0.58 mg/L) (Table 1). In contrast to the study sites, there was no significant difference between monthly averages of total N contents of the precipitation and total N values varied between 0.44 and 0.88 mg/L (Table 2). Total N had significant correlation coefficients only with TSP, Ca hardness, Ca^{2+}, and organic matter for the precipitation at the forest site (Table 3) and with Ca hardness and Ca^{2+} for the rainwater at the forest village (Table 4).

In addition to HCO_3^-, NO_3^- was another anion that differed significantly with respect to the study sites ($P < 0.001$), and the forest village had a higher NO_3^- concentration in the rainwater compared to the forest site (Table 1). The overall average monthly NO_3^- values varied between 1.08 and 5.87 mg/L and did not show significant changes throughout the year (Table 2). The concentration of NO_3^- had significant correlation coefficients with Na^+ ($r = 0.35$, $P < 0.05$), PO_4^{3-} ($r = -0.51$, $P < 0.01$), Fe ($r = 0.58$, $P < 0.01$), Al ($r = -0.33$, $P < 0.05$), and NH_4^+ ($r = 0.49$, $P < 0.01$) only for the precipitation at the forest site (Table 3).

Concentration of PO_4^{3-} in the precipitation was similar between study sites and was 26.98 mg/L for the forest and 23.13 mg/L for the forest village (Table 1). Mean monthly PO_4^{3-} values did not show significant differences among the months and varied between 17.83 and 34.15 mg/L (Table 2). PO_4^{3-} had significant correlations with TSP, Cl^-, organic matter, NO_3^-, Al, and SO_4^{2-} for the rainwater at the forest site (Table 3) while it had significant correlation only with Fe for the rainwater at the forest village site (Table 4).

The concentration of Fe ion in the rainwater did not show significant changes with respect to study sites and were 222.73 μg/L and 245.63 μg/L for the forest and forest village study sites, respectively (Table 1). In contrast, overall average monthly Fe values significantly differed among the months and reached the highest value in February (495.25 μg/L) and the lowest value (52.71 μg/L) in June (Fig. 1d). Fe ion had significant correlations with turbidity and NO_3^- for the forest site (Table 3) and EC, TSP, organic matter, and PO_4^{3-} for the forest village site (Table 4). Precipitation at the forest and forest village sites had similar amount of Al ions with low concentrations (Table 1) and the overall average Al

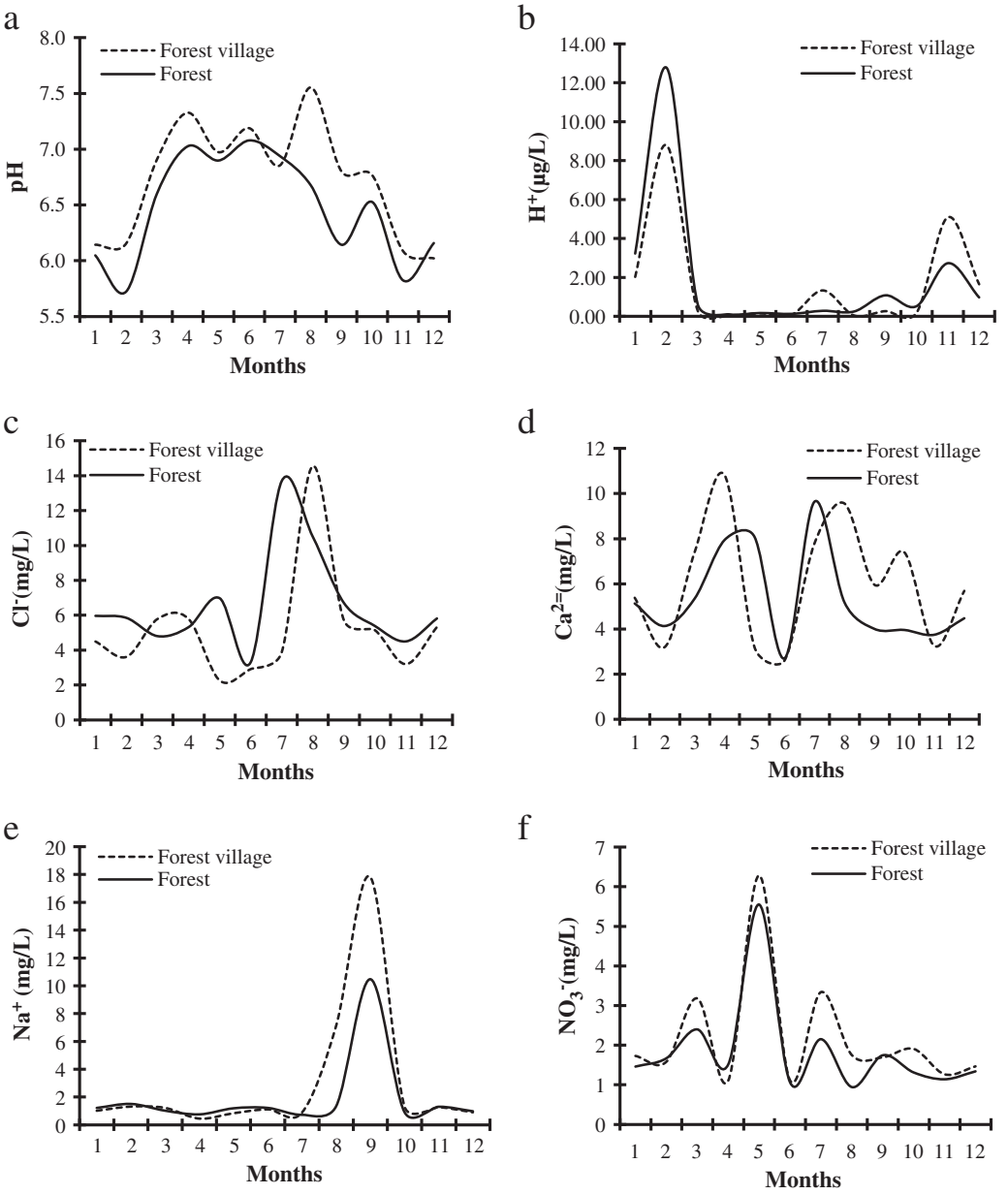


Fig. 3. Average monthly concentrations of some ions in the precipitation at the forest and forest village study sites.

concentrations did not show significant changes throughout the year (Table 2). Al had significant correlation coefficients with TSP, Mg^{2+}, NO_3^-, PO_4^{3-}, and SO_4^{2-} for the precipitation at the forest site (Table 3) whereas it had significant correlations with EC and SO_4^{2-} at the forest village site (Table 4).

The concentration of NH_4^+ in the precipitation was also significantly higher in the forest site (0.39 mg/L) compared to the forest village (0.37 mg/L) (Table 1). The concentration of NH_4^+ changed significantly throughout the year and ranged from 0.22 mg/L in November to 0.75 mg/L in May (Fig. 1e). Similar to Fe, the concentration of NH_4^+ decreased in the second half of the year (Fig. 1e). NH_4^+ had significant correlation coefficients with total alkalinity ($CaCO_3$), HCO_3^-, K^+, and NO_3^- for the precipitation at the forest site (Table 3) and with EC and SO_4^{2-} for the rainwater at the forest village (Table 4).

The concentration of SO_4^{2-} was similar for both forest and forest village sites and did not change significantly throughout the year (Table 2). The overall average SO_4^{2-} values averaging over the months were 7.69 mg/L and 6.78 mg/L for the forest and the forest village sites, respectively (Table 1). On the other hand, overall mean monthly SO_4^{2-} values averaged over study sites did not show significant changes and varied between 3.29 and 15.92 mg/L (Table 2). The concentration of SO_4^{2-} had

Table 2
Overall average monthly values (mean ± SEM) of rainwater quality parameters.

Parameters	Months												Probability
	1	2	3	4	5	6	7	8	9	10	11	12	
Turbidity(NTU)	4.74±0.63	9.38±2.73	17.83±5.30	8.44±2.08	24.51±12.06	8.76±2.01	11.87±3.01	9.18±1.98	7.14±1.04	19.33±16.18	18.59±14.99	4.57±2.01	$P > 0.05$
TSP (mg/L)	0.04±0.01	0.04±0.01	0.06±0.01	0.05±0.02	0.06±0.02	0.04±0.01	0.05±0.01	0.06±0.02	0.03±0.01	0.02±0.003	0.03±0.004	0.04±0.02	$P > 0.05$
Cl^- (mg/L)	5.30±0.74	4.95±0.92	5.25±0.91	5.47±1.51	5.21±1.96	3.17±0.69	9.77±5.18	11.51±2.38	6.22±1.30	5.26±0.71	3.85±0.85	5.59±0.84	$P > 0.05$
Total hardness($CaCO_3$) (mg/L)	46.40±6.35	38.82±4.06	44.12±5.56	57.83±6.91	59.41±7.97	29.75±6.81	45.16±10.71	45.70±5.46	35.97±2.77	48.20±8.53	35.86±7.33	43.02±6.25	$P > 0.05$
Ca hardness($CaCO_3$) (mg/L)	12.78±2.76	9.31±2.75	15.46±1.98	22.50±2.89	15.40±3.51	6.55±1.12	21.99±6.36	15.43±3.57	12.34±2.60	13.94±5.20	8.60±0.87	12.45±3.48	$P > 0.05$
Ca^{2+} (mg/L)	5.25±1.09	3.76±1.10	6.28±0.79	8.90±1.12	6.24±1.40	2.67±0.43	8.90±2.54	6.28±1.47	4.98±1.04	5.68±2.08	3.50±0.35	5.03±1.39	$P > 0.05$
Mg^{2+} (mg/L)	8.13±1.10	7.38±0.70	6.99±1.05	8.73±1.55	10.74±2.04	5.66±1.39	5.65±1.28	7.39±0.48	5.77±0.56	8.33±1.54	6.65±1.67	7.52±1.28	$P > 0.05$
Organic matter (mg/L)	2.63±0.38	3.55±1.24	2.62±0.41	4.11±0.94	5.52±2.14	2.51±0.97	5.47±2.41	7.52±1.92	2.47±0.52	2.05±0.32	2.30±0.55	2.67±0.54	$P > 0.05$
Total N (mg/L)	0.59±0.12	0.61±0.13	0.57±0.08	0.88±0.35	0.69±0.10	0.52±0.05	0.65±0.18	0.68±0.05	0.48±0.06	0.73±0.22	0.69±0.22	0.44±0.06	$P > 0.05$
Na^+ (mg/L)	1.12±0.16	1.42±0.25	1.12±0.16	0.65±0.25	1.05±0.24	1.18±0.48	0.80±0.15	3.26±1.94	14.14±9.02	1.11±0.27	1.29±0.28	0.94±0.19	$P > 0.05$
K^+ (mg/L)	0.91±0.15	0.95±0.22	0.87±0.13	1.21±0.75	1.64±0.69	0.20±0.01	1.07±0.06	1.91±0.75	0.62±0.15	1.21±0.31	0.76±0.14	0.64±0.06	$P > 0.05$
NO_3^- (mg/L)	1.60±0.35	1.63±0.35	2.79±0.47	1.35±0.33	5.87±1.51	1.08±0.43	2.75±0.66	1.21±0.28	1.72±0.72	1.61±0.49	1.20±0.42	1.40±0.43	$P > 0.05$
PO_4^{3-} (mg/L)	17.83±4.90	21.86±6.14	18.95±3.77	21.78±1.45	19.43±4.68	20.84±12.73	25.07±10.18	30.83±4.72	34.15±6.92	30.34±4.90	30.87±6.20	26.83±5.00	$P > 0.05$
Al (μg/L)	82.68±37.99	46.58±30.85	98.87±55.74	106.74±56.38	125.38±68.14	96.94±72.65	15.80±6.34	89.77±64.73	140.41±57.19	56.29±25.10	71.04±38.02	44.39±25.33	$P > 0.05$
SO_4^{2-} (mg/L)	8.98±2.98	7.38±3.22	9.53±2.28	8.76±3.17	7.55±2.79	3.29±2.45	6.51±3.37	15.92±5.18	5.45±1.52	5.41±1.16	5.71±1.19	5.94±1.82	$P > 0.05$

significant correlations with pH, EC, turbidity, TSP, total alkalinity ($CaCO_3$), HCO_3^-, Cl^-, total hardness ($CaCO_3$), Ca hardness, Ca^{2+}, organic matter, K^+, PO_4^{3-}, and Al in the forest study site (Table 3) and with EC, TSP, total alkalinity ($CaCO_3$), HCO_3^-, Cl^-, Ca hardness, Ca^{2+}, organic matter, Al, and NH_4^+ in the forest village study site (Table 4).

4. Discussion

The values of rainwater quality parameters measured in this study were relatively higher but rainwater pH was lower than those found by Migliavacca et al. (2004) and Song and Gao (2009). Na^+ and Cl^- concentrations in the precipitation at the forest and forest village sites were higher and Mg^{2+} and NO_3^- concentrations were lower than those in the precipitation at central Scotland (Miller et al., 2000). Additionally, the order of the ions in the present study was different from those reported for some other areas. For instance, SO_4^{2-} and H^+ were the dominant anion and cation in the precipitation at a study site in Singapore (Hu et al., 2003); SO_4^{2-} and Ca^{2+} at northeast Spain (Alastuey et al., 1999), Kaynarca and Istanbul, Turkey (Okay et al., 2002; Akkoyunlu and Tayanç, 2003), India (Mouli et al., 2005), and China (Tu et al., 2005); SO_4^{2-} and Al at Istanbul, Turkey (Başak and Alagha, 2004); SO_4^{2-} and K^+ at Belgium (Staelens et al., 2005); SO_4^{2-} and NH_4^+ at Ankara, Turkey (Topçu et al., 2002) and Newark (Song and Gao, 2009); and Cl^- and Ca^{2+} at Lhasa (Zhang et al., 2003). The average values of some ions (Cl^-, NO_3^-, Ca^{2+}, SO_4^{2-}, Mg^{2+}, Na^+, Al, Fe, and K^+) in this study were much higher than those found in other studies conducted by Okay et al. (2002) and Akkoyunlu and Tayanç (2003) but some (NO_3^-, NH_4^+, SO_4^{2-}) were lower than those found by Gülsoy et al. (1999) in Istanbul. Compared to a study conducted in Istanbul (Başak and Alagha, 2004), rainwater at both forest and forest village study sites had lower Cl^-, NO_3^-, Ca^{2+}, SO_4^{2-}, and K^+ concentrations but higher NH_4^+, Mg^{2+}, Al, and Fe values. As understood from that information, it is hard to make generalizations about atmospheric environmental quality of Istanbul since the results of different studies were not consistent with each other. Concentrations of the ions in the present study were also much higher (except for NH_4^+) than those reported in a study conducted in Tibet (Zhang et al., 2003) but higher than all ions studied in the east coast of India (Das et al., 2005). The average pH values of both forest and forest village study sites were higher than that found by Başak and Alagha (2004) but lower than that reported by Okay et al. (2002) for the precipitation at Istanbul. Result of the present study was consistent with findings of Alastuey et al. (1999), Topçu et al. (2002), Zhang et al. (2003), Safai et al. (2004), and Bayraktar and Turalioglu (2005) that pH values of the precipitation was alkaline in contrast to findings of Lee et al. (2000), Başak and Alagha (2004), Hu et al. (2003), Migliavacca et al. (2004), Tu et al. (2005), and Song and Gao (2009). The alkaline character of precipitation can be considered as an indicator of low H^+ concentration. In contrast to findings from some other studies, significant correlations were not found between H^+ and major acidifying anions (NO_3^- and SO_4^{2-}) for the precipitation at both forest and forest village sites similar to the results reported for the precipitation at Istanbul (Başak and Alagha, 2004), Pune, India (Safai et al., 2004), and Eastern China (Cheng et al., 2011).

Table 3
Correlation coefficients between rainwater quality parameters for the forest study site.

	pH	H^+	EC	Turbidity	TSP	Total alkalinity ($CaCO_3$)	HCO_3^-	Cl^-	Total hardness ($CaCO_3$)	Ca hardness	Ca^{2+}	Mg^{2+}	Organic matter	Total N	Na^+	K^+	NO_3^-	PO_4^{3-}	Fe	Al	NH_4^+	SO_4^{2-}
pH	1	**−0.59**[a]	**0.50**[a]	0.22	**0.32**[b]	**0.41**[a]	**0.48**[a]	**0.31**[b]	**0.41**[a]	**0.57**[a]	**0.57**[a]	0.13	**0.44**[a]	0.13	−0.20	0.14	0.10	0.08	−0.01	0.06	0.14	**0.46**[a]
H^+		1	−0.16	−0.08	−0.20	−0.16	−0.17	−0.14	−0.12	−0.21	−0.21	−0.01	−0.17	−0.16	0.00	−0.04	0.12	−0.16	0.27	−0.03	−0.06	−0.18
EC			1	**0.53**[a]	**0.46**[a]	**0.38**[a]	**0.54**[a]	**0.51**[a]	**0.42**[a]	**0.61**[a]	**0.61**[a]	0.11	**0.54**[a]	0.24	−0.18	0.28	0.29	0.21	0.28	0.09	0.06	**0.54**[a]
Turbidity				1	**0.49**[a]	**0.39**[a]	**0.47**[a]	0.24	0.21	**0.50**[a]	**0.51**[a]	−0.09	**0.48**[a]	0.07	0.02	0.29	0.27	−0.10	**0.52**[a]	−0.08	0.15	**0.36**[b]
TSP					1	**0.54**[a]	**0.58**[a]	**0.43**[a]	0.27	**0.71**[a]	**0.71**[a]	−0.05	**0.58**[a]	**0.38**[b]	−0.17	**0.34**[b]	0.01	**0.30**b[b]	0.20	**0.36**[b]	0.08	**0.75**[a]
Total alkalinity ($CaCO_3$)						1	0.84[a]	**0.41**[a]	**0.36**[b]	**0.52**[a]	**0.52**[a]	0.10	**0.41**[a]	0.25	−0.12	0.14	0.26	0.23	0.15	0.25	**0.59**[a]	**0.45**[a]
HCO_3^-							1	**0.51**[a]	**0.36**[a]	**0.58**[a]	**0.58**[a]	0.07	**0.59**[a]	0.17	−0.11	**0.62**[a]	0.24	0.28	0.01	0.09	**0.52**[a]	**0.54**[a]
Cl^-								1	**0.44**[a]	**0.68**[a]	**0.68**[a]	0.09	**0.73**[a]	0.20	−0.21	**0.54**[a]	−0.15	**0.59**[a]	−0.13	0.04	0.23	**0.54**[a]
Total hardness ($CaCO_3$)									1	**0.66**[a]	**0.67**[a]	**0.85**[a]	**0.50**[a]	0.17	−0.24	0.15	0.00	0.02	−0.07	−0.28	0.22	**0.42**[a]
Ca hardness										1	**1.00**[a]	0.16	**0.78**[a]	**0.38**[b]	−0.13	**0.38**[b]	0.03	0.24	0.14	0.13	0.19	**0.74**[a]
Ca^{2+}											1	0.17	**0.78**[a]	**0.35**[b]	−0.13	**0.39**[b]	0.03	0.25	0.13	0.12	0.19	**0.74**[a]
Mg^{2+}												1	0.11	0.04	−0.24	−0.09	−0.04	−0.11	−0.15	**−0.43**[a]	0.16	0.06
Organic matter													1	**0.39**[b]	−0.15	**0.62**[a]	0.01	**0.54**[a]	0.09	0.01	0.24	**0.67**[a]
Total N														1	−0.08	−0.16	−0.09	0.14	0.13	0.23	0.11	0.25
Na^+															1	0.32	**0.35**[b]	−0.23	−0.02	−0.11	−0.04	−0.19
K^+																1	0.23	−0.07	0.08	−0.06	**0.35**[b]	**0.32**[b]
NO_3^-																	1	**−0.51**[a]	**0.58**[a]	**−0.33**[b]	**0.49**[a]	−0.05
PO_4^{3-}																		1	−0.25	**0.46**[a]	−0.24	**0.37**[b]
Fe																			1	0.21	0.16	−0.01
Al																				1	−0.18	**0.47**[a]
NH_4^+																					1	0.11
SO_4^{2-}																						1

Bold numbers show statistically significant correlation coefficients between rainwater parameters.
[a] Significant at α level of 0.01.
[b] Significant at α level of 0.05.

Table 4
Correlation coefficients between rainwater quality parameters for the forest village study site.

	pH	H^+	EC	Turbidity	TSP	Total alkalinity ($CaCO_3$)	HCO_3^-	Cl^-	Total hardness ($CaCO_3$)	Ca hardness	Ca^{2+}	Mg^{2+}	Organic matter	Total N	Na^+	K^+	NO_3^-	PO_4^{3-}	Fe	Al	NH_4^+	SO_4^{2-}
pH	1	**−0.65**[a]	**0.39**[a]	0.07	0.14	**0.47**[a]	**0.47**[a]	**0.41**[a]	0.30	**0.34**[b]	**0.33**[b]	0.17	0.21	−0.02	0.03	0.33	0.15	−0.13	0.22	0.15	0.06	0.26
H^+		1	−0.11	−0.07	−0.15	−0.20	−0.20	−0.29	**−0.32**[b]	−0.23	−0.23	−0.25	−0.12	−0.13	−0.05	−0.18	−0.15	0.28	−0.05	0.16	−0.08	0.04
EC			1	0.20	**0.53**[a]	0.28	0.28	**0.68**[a]	**0.31**[b]	**0.60**[a]	**0.59**[a]	0.03	**0.52**[a]	0.01	−0.20	0.10	−0.16	0.09	**0.45**[a]	**0.38**[b]	**0.51**[a]	**0.66**[a]
Turbidity				1	0.18	−0.13	−0.13	−0.12	−0.05	−0.06	−0.06	−0.03	−0.04	0.30	−0.05	−0.03	−0.07	−0.16	0.09	−0.11	−0.08	−0.01
TSP					1	**0.38**[b]	**0.38**[b]	**0.48**[a]	0.20	0.25	0.26	0.11	**0.58**[a]	0.05	−0.17	0.24	0.07	−0.08	**0.56**[a]	0.05	0.33	**0.54**[a]
Total alkalinity ($CaCO_3$)						1	**1.00**[a]	**0.34**[b]	**0.37**[b]	**0.35**[b]	**0.35**[b]	0.25	**0.43**[a]	−0.05	−0.01	0.29	−0.04	0.08	0.21	0.24	−0.09	**0.43**[a]
HCO_3^-							1	**0.34**[b]	**0.37**[b]	**0.35**[b]	**0.35**[b]	0.25	**0.43**[a]	−0.05	−0.01	0.29	−0.04	0.08	0.21	0.24	−0.09	**0.43**[a]
Cl^-								1	0.28	**0.54**[a]	**0.54**[a]	0.03	**0.50**[a]	−0.02	−0.11	**0.36**[b]	−0.18	−0.04	0.21	0.30	0.05	**0.61**[a]
Total hardness ($CaCO_3$)									1	**0.60**[a]	**0.60**[a]	**0.88**[a]	0.26	0.28	−0.14	−0.10	−0.10	−0.13	0.07	−0.06	0.06	0.19
Ca hardness										1	**1.00**[a]	0.15	**0.41**[a]	**0.43**[b]	−0.01	0.05	−0.15	0.01	0.08	0.23	0.11	**0.42**[a]
Ca^{2+}											1	0.15	**0.42**[a]	**0.43**[b]	−0.01	0.05	−0.15	0.00	0.08	0.22	0.11	**0.42**[a]
Mg^{2+}												1	0.08	0.08	−0.16	−0.15	−0.03	−0.15	0.04	−0.19	0.00	−0.02
Organic matter													1	0.04	−0.19	0.30	−0.11	0.00	**0.36**[b]	0.11	0.21	**0.55**[a]
Total N														1	−0.10	−0.23	−0.11	−0.03	−0.12	0.08	−0.05	0.17
Na^+															1	**0.52**[a]	0.24	−0.17	−0.07	−0.07	−0.14	−0.11
K^+																1	0.12	−0.10	0.35	0.04	−0.30	0.29
NO_3^-																	1	−0.27	0.22	−0.02	0.09	−0.13
PO_4^{3-}																		1	**−0.34**[b]	0.29	0.11	0.26
Fe																			1	0.13	0.27	0.29
Al																				1	0.05	**0.52**[a]
NH_4^+																					1	**0.31**[b]
SO_4^{2-}																						1

Bold numbers show statistically significant correlation coefficients between rainwater parameters.
[a] Significant at α level of 0.01.
[b] Significant at α level of 0.05.

Although the precipitation at both forest and forest village study sites had higher EC values compared to findings from other studies (Topçu et al., 2002; Zhang et al., 2003; Migliavacca et al., 2004; Yi et al., 2010; Cheng et al., 2011), EC values could be still considered as low values and hence, an indicator of clean atmospheric environment in Belgrad Forest (Cheng et al., 2011). As for the rainwater quality parameters, besides low EC values, low turbidity, TSP, and organic matter values also can be evaluated as an indicator of good atmospheric environmental quality in the study sites.

High correlation between TSP and SO_4^{2-} showed that the total suspended particles influenced acidity of the precipitation at the study sites as found by Zhang et al. (2003) and Song and Gao (2009). In an 18-year long monitoring study in the same study site, Gökbulak et al. (2008) found that average monthly HCO_3^- concentrations in the bulk precipitation varied between 5.09 and 17.96 mg/L and in general, HCO_3^- concentration decreased in the winter months as found in the present study. Also, high correlations between HCO_3^- and both K^+ and Ca^{2+} cations can be an indicator of the formation of calcium and potassium carbonates and therefore, HCO_3^- could be considered as one of the ions that neutralized SO_4^{2-} in the study site (Safai et al., 2004).

Compared to the results of some studies carried out in Istanbul and other parts of Turkey, the concentration of Cl^- in the present study was lower than that reported by Başak and Alagha (2004) but higher than that found by Okay et al. (2002), Topçu et al. (2002), and Gökbulak et al. (2008). The concentration of Na^+ in the present study was found to be higher than those reported in other studies conducted in the same and different regions around the world (Okay et al., 2002; Topçu et al., 2002; Zhang et al., 2003; Başak and Alagha, 2004; Das et al., 2005). The results of this study were not consistent with the findings of Okay et al. (2002), Safai et al. (2004), Başak and Alagha (2004), Mouli et al. (2005), and Cheng et al. (2011) that high correlations were found between Na^+ and other ions including sea origin Cl^- ion. These results indicated that most of the ions in the precipitation at the study sites could be most probably derived from the same crustal and anthropogenic sources (Tu et al., 2005). Due to the short distance between the Black Sea and the study sites (about 7 km), prevailing wind direction from NW, and rough topography, sea breeze could be blocked and this case would have been caused deposition of the sea-originated Na^+ and Cl^- in the soil (Migliavacca et al., 2004; Cheng et al., 2011).

The average monthly concentrations of Ca^{2+} and Mg^{2+} in the present study were higher but the concentration of K^+ was lower than those reported in a study conducted in the same forest site (Gökbulak et al., 2008). Moreover, the concentrations of Ca^{2+}, Mg^{2+}, and K^+ were found to be higher than those found in Istanbul and other parts of Turkey (Okay et al., 2002; Topçu et al., 2002; Bayraktar and Turalioglu, 2005) while the concentration of Ca^{2+} was lower than that found by Başak and Alagha (2004) for Istanbul. Similarly, the concentrations of Ca^{2+}, Mg^{2+}, and K^+ ions in the present study were higher than those in other works carried out in Tibet (Zhang et al., 2003), India (Das et al., 2005), the US East Coast (Song and Gao, 2009), northeast Spain (Alastuey et al., 1999), and Singapore (Hu et al., 2003). Based on the results of studies conducted in Istanbul, it seemed that the concentrations of the base cations in the precipitation at Belgrad Forest were generally higher than those at other parts of the city. This case can be attributed to the sampling type of the precipitation because Staelens et al. (2005) suggested that Ca^{2+}, Mg^{2+}, and K^+ ions were often higher in bulk precipitation samples than wet-only samples due to deposition of these soil-derived cations in the collectors.

The results of the present study for the total N content were not consistent with the findings of other studies. For instance, compared to total N content of the precipitation in the present study, a higher N concentration was found in the precipitation in a study conducted about two decades ago at the same region (Gökbulak et al., 2008). Similarly, even though Miller et al. (2000) measured NH_4–N and NO_3–N, not total N, between 1989 and 1998 years in central Scotland they found a higher N content in the rainfall than that measured in the present study. This difference can be attributed to study site conditions during the study period such as climate, topography, land use types, agricultural activities, and sampling season and intervals (Heuer et al., 2000; Kang et al., 2004; Cheng et al., 2011).

On the other hand, highly significant correlations between NO_3^- by NH_4^+ can explain neutralization of NO_3^- by NH_4^+ and high pH value for the precipitation at the forest site (Topçu et al., 2002; Safai et al., 2004; Bayraktar and Turalioglu, 2005; Mouli et al., 2005). The result of this study was not consistent with findings of other studies conducted in Istanbul and other parts of Turkey. Average NO_3^- concentration in the present study was within the range of nitrate concentrations reported by Başak and Alagha (2004) but higher than those found by Okay et al. (2002) and Akkoyunlu and Tayanç (2003), and lower than that measured by Gülsoy et al. (1999) for Istanbul. Also, the concentration of NO_3^- in the current study was found to be higher than those reported in other works done in different parts of Turkey (Kaya and Tuncel, 1997; Topçu et al., 2002; Bayraktar and Turalioglu, 2005) and different from those found in other studies around the world (Heuer et al., 2000; Zhang et al., 2003; Das et al., 2005; Mouli et al., 2005; Staelens et al., 2005; Song and Gao, 2009; Yi et al., 2010; Cheng et al., 2011). Additionally, the average monthly PO_4^{3-} content of the precipitation in the present study was higher than those measured in the same study area (Gökbulak et al., 2008) and in India (Das et al., 2005) and Uruguay (Zunckel et al., 2003).

Compared to other ions, Fe concentration of the precipitation was analyzed in a few studies and concentrations of Fe ion in these studies were found to be lower than that found in the present study (Kaya and Tuncel, 1997; Başak and Alagha, 2004; Herrera et al., 2009; Song and Gao, 2009; Cheng et al., 2011). On the other hand, compared to the present study, lower Al concentrations were reported in other studies conducted in China (Cheng et al., 2011), Istanbul, Turkey (Kaya and Tuncel, 1997; Başak and Alagha, 2004), US East coast (Song and Gao, 2009), South Brazil (Migliavacca et al., 2004), and Central Scotland (Miller et al., 2000) and a higher Al concentration in Costa Rica (Herrera et al., 2009). The precipitation at Belgrad Forest had a lower NH_4^+ concentration than most places studied in Turkey and other regions around the world (Gülsoy et al., 1999; Miller et al., 2000; Akkoyunlu and Tayanç, 2003; Kang et al., 2004; Staelens et al., 2005; Herrera et al., 2009; Song and Gao, 2009; Cheng et al., 2011).

Table 5
Correlation coefficients between forest and forest village study sites for the same rainwater quality parameters.

	pH	H^+	EC	Turbidity	TSP	Total alkalinity ($CaCO_3$)	HCO_3^-	Cl^-	Total hardness ($CaCO_3$)	Ca hardness	Ca^{2+}	Mg^{2+}	Organic matter	TotalN	Na^+	K^+	NO_3^-	PO_4^{3-}	Fe	Al	NH_4^+	SO_4^{2-}
pH	**0.74**[a]	**−0.58**[b]	0.25	0.25	0,22	**0.35**[b]	**0.37**[b]	0.30	0.25	0.29	0.29	0.15	0.28	0.14	0.03	0.04	−0.00	0.05	−0.06	0.02	0.16	0.19
H^+	**−0.59**[a]	**0.92**[a]	−0.17	−0.09	−0.20	−0.17	−0.18	−0.17	−0.20	−0.20	−0.20	−0.14	−0.22	−0.06	−0.04	0.02	0.06	−0.10	0.22	0.14	−0.05	−0.12
EC	0.12	−0.08	**0.35**[b]	**0.60**[a]	**0.45**[a]	0.12	0.11	**0.72**[a]	0.11	0.21	0.20	0.01	**0.40**[b]	0.19	−0.13	0.18	−0.174	0.23	0.10	0.23	0.01	**0.41**[a]
Turbidity	0.03	−0.07	0.02	**0.34**[b]	0.01	−0.09	−0.09	−0.19	−0.07	−0.06	−0.06	−0.05	−0.05	−0.15	−0.03	−0.10	−0.132	−0.08	0.01	−0.17	−0.10	−0.06
TSP	0.22	−0.21	−0.07	**0.39**[b]	**0.48**[a]	0.24	0.23	0.27	0.06	0.25	0.24	0.03	0.17	0.08	−0.19	−0.12	−0.14	0.08	0.16	−0.06	0.10	−0.01
Total alkalinity ($CaCO_3$)	**0.37**[b]	−0.26	−0.01	**−0.02**	0.30	**0.66**[a]	**0.60**[a]	0.01	0.32	0.24	0.24	0.26	0.11	0.13	−0.04	−0.02	0.10	0.11	−0.06	0.12	**0.46**[a]	0.19
HCO_3^-	**0.37**[b]	−0.26	−0.01	**−0.02**	0.30	**0.66**[a]	**0.60**[a]	0.01	0.32	0.24	0.24	0.26	0.11	0.13	−0.04	−0.02	0.10	0.11	−0.06	0.12	**0.46**[a]	0.19
Cl^-	0.26	−0.25	0.02	0.04	0.06	0.28	0.24	**0.41**[b]	0.05	0.13	0.13	−0.02	0.15	0.19	−0.15	0.09	−0.19	0.23	0.07	0.18	0.03	0.23
Total hardness ($CaCO_3$)	**0.42**[a]	**−0.37**[b]	0.04	0.04	0.12	**0.37**[b]	0.27	0.15	**0.68**[a]	0.30	0.30	**0.69**[a]	0.07	**0.38**[b]	−0.16	−0.25	−0.03	0.09	−0.16	−0.04	0.26	0.12
Ca Hardness	0.24	−0.21	0.02	−0.02	0.09	0.18	0.19	0.25	**0.36**[b]	0.29	0.29	0.29	0.13	**0.66**[a]	−0.05	−0.23	−0.23	0.29	−0.21	0.19	−0.01	0.25
Ca^{2+}	0.23	−0.21	0.01	−0.02	0.08	0.18	0.18	0.24	**0.36**[b]	0.28	0.28	0.29	0.12	**0.66**[a]	−0.05	−0.23	−0.23	0.28	−0.21	0.18	−0.01	0.24
Mg^{2+}	**0.39**[b]	**−0.33**[b]	0.03	0.06	0.10	**0.35**[b]	0.23	0.04	**0.62**[a]	0.20	0.20	**0.67**[a]	0.01	0.08	−0.17	−0.17	0.11	−0.06	−0.08	−0.17	**0.32**[b]	−0.00
Organic matter	0.31	−0.19	0.01	**0.34**[b]	**0.41**[b]	**0.48**[a]	**0.41**[a]	0.25	0.22	**0.45**[a]	**0.45**[a]	0.03	**0.36**[b]	0.11	−0.22	−0.05	−0.14	0.18	−0.06	0.16	0.26	**0.39**[b]
Total N	0.02	−0.19	0.02	**−0.10**	0.26	0.06	−0.01	0.07	0.18	0.15	0.14	0.15	0.19	**0.51**[a]	−0.11	−0.29	−0.15	0.04	−0.19	0.05	0.08	0.14
Na^+	−0.24	0.08	−0.18	**0.02**	−0.18	−0.12	−0.12	−0.27	−0.24	−0.14	−0.13	−0.23	−0.18	−0.04	**1.00**[a]	−0.02	**0.34**[b]	−0.20	−0.04	−0.11	−0.05	−0.18
K^+	0.12	−0.09	−0.17	−0.17	−0.14	0.09	0.11	−0.14	−0.22	−0.28	−0.27	−0.14	−0.16	−0.07	**0.57**[a]	**0.60**[a]	0.20	−0.15	0.12	−0.12	0.27	−0.10
NO_3^-	0.28	−0.01	**0.48**[a]	**0.39**[b]	0.16	0.07	0.06	−0.26	0,00	0.08	0.08	−0.06	−0.05	0.05	0.25	0.04	**0.85**[a]	**−0.43**[a]	**0.67**[a]	**−0.34**[b]	0.11	0,00
PO_4^{3-}	−0.07	−0.01	0.19	−0.15	0.20	−0.06	0.05	**0.39**[b]	−0.16	0.08	0.08	−0.24	**0.40**[b]	0.29	−0.19	0.05	**−0.40**[b]	**0.70**[a]	−0.20	**0.43**[b]	−0.28	0.31
Fe	0.18	0.05	−0.02	**0.50**[a]	0.24	0.33	0.25	−0.02	0.13	0.17	0.17	0.07	−0.08	−0.13	−0.06	0.28	0.22	−0.15	**0.59**[a]	0.13	0.28	0.12
Al	0.13	0.00	0.15	−0.08	0.31	0.21	0.12	0.18	0.04	0.28	0.28	−0.08	0.19	0.12	−0.08	0.09	−0.06	**0.33**[b]	0.05	**0.87**[a]	−0.09	**0.56**[a]
NH_4^+	0.18	−0.07	**0.43**[a]	**0.42**[b]	**0.39**[b]	0.11	0.13	**0.43**[a]	**0.37**[b]	**0.45**[a]	**0.45**[a]	0.22	**0.34**[b]	0.26	−0.13	−0.14	−0.08	0.15	0.22	0.11	0.01	**0.45**[a]
SO_4^{2-}	**0.34**[b]	−0.15	0.23	0.05	**0.45**[a]	**0.49**[a]	**0.48**[a]	**0.50**[a]	0.24	**0.50**[a]	**0.50**[a]	0.02	0.32	0.27	−0.15	0.01	−0.20	**0.37**[b]	0.01	**0.37**[b]	0.06	**0.66**[a]

Bold numbers show statistically significant correlation coefficients between rainwater parameters.
[a] Significant at α level of 0.01.
[b] Significant at α level of 0.05.

Significant correlations between $(NH_4)^+$ and NO_3^- and between SO_4^{2-} and Ca^{2+} at the forest study site and between SO_4^{2-} and NH_4^+ at the forest village indicated that precipitation acidity was also neutralized by NH_4^+ in addition to Ca^{2+} because NO_3^-, SO_4^{2-}, NH_4^+, and Ca^{2+} are the major ions affecting acidity of precipitation and its neutralization (Song and Gao, 2009). Thus, high pH values in the present study can be attributed to the neutralization of precipitation by NH_4^+ and Ca^{2+} ions.

SO_4^{2-} values found in the present study were higher than those reported for Ankara (Kaya and Tuncel, 1997; Topçu et al., 2002) but lower than those for Istanbul (Gülsoy et al., 1999; Okay et al., 2002; Başak and Alagha, 2004). However, Akkoyunlu and Tayanç (2003) found a higher SO_4^{2-} concentration in the alkaline but lower in the acidic precipitation at Istanbul. Moreover, the results showed that the precipitation at Belgrad Forest of Istanbul had a higher SO_4^{2-} concentration than those in most of the other places around the world including India (Safai et al., 2004; Mouli et al., 2005), Belgium (Staelens et al., 2005), Korea (Lee et al., 2000; Kang et al., 2004), Uruguay (Zunckel et al., 2003), Spain (Alastuey et al., 1999), Singapore (Hu et al., 2003), Tibet (Zhang et al., 2003), USA (Heuer et al., 2000; Song and Gao, 2009), Japan (Sakihama et al., 2008), and Brazil (Migliavacca et al., 2004) but lower than in China (Tu et al., 2005; Yi et al., 2010).

A lower pH and higher turbidity, TSP, organic matter, Fe, and SO_4^{2-} values were expected to be found in the precipitation at the forest village because of motor vehicle traffic, fossil fuel consumption, and presence of bare soil surface, but significant differences were not determined between study sites for these and some other parameters (Table 1). Despite motor vehicle traffic and fossil fuel consumption in the forest village, high pH values were found for both study sites due to neutralization of SO_4^{2-} and NO_3^- by NH_4^+ and Ca^{2+} (Topçu et al., 2002; Safai et al., 2004; Mouli et al., 2005). Additionally, significant correlation coefficients were found between study sites for the same precipitation parameters except for Ca hardness, Ca^{2+}, and NH_4^+ (Table 5). This meant that both study sites were under the influence of the same emission sources. Based on the results of this and some other studies conducted in the same region, it is hard to make generalization about trend of chemical composition of the precipitation and atmospheric air quality because the results of these studies were not consistent with each other. For instance, we found higher or lower ion concentrations than some researchers did (Gülsoy et al., 1999; Okay et al., 2002; Akkoyunlu and Tayanç, 2003; Başak and Alagha, 2004). Differences in the results of these studies can be attributed to differences in the sampling type (bulk deposition or wet-only deposition), sampling interval (periodically or event-based), sampling season and period, and physical characteristics of study sites. Moreover, other studies were carried out in settlement areas in addition to rural areas with different population density, industrial facilities, motor vehicle traffic, agricultural activities, sampling interval, and season compared to this study.

5. Conclusion

In this study, chemical compositions of the precipitations were characterized in Belgrad Forest and a forest village, Bahçeköy, located in the same forest ecosystem and chemical contents of the precipitations at both study sites were compared in order to see if settlements in the forest ecosystems threaten forest ecosystem health by causing damages to atmospheric air quality. The results showed that the concentration of SO_4^{2-} in the precipitation at Belgrad forest was very high compared to those found in different areas around the world. The precipitation was slightly acidic during the winter months and acidic agents (SO_4^{2-} and NO_3^-) were neutralized by NH_4^+ Ca^{2+}. Despite the human originated activities such as vehicle traffic and fossil fuel consumption for cooking and heating in the forest village, there were no big differences between precipitation chemistries of the forest and forest village due to absence of industrial activities in the forest village. Significant correlations found between study sites for the same rainwater quality parameters also indicated that both study sites were under the influence of the same emission sources which would be either from the forest village or from other sources. Therefore, further studies are needed to determine emission sources for the region. Moreover, the concentrations of some rainwater parameters did not show significant changes throughout the year. It can be concluded that the unindustrialized residential areas in the forest ecosystems may not be a very serious threat at the moment for the rainwater quality and contamination of water resources and hence forest ecosystem health in the forestlands.

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