EFFECTS OF ATMOSPHERIC DEPOSITION ON SOME PROCESSES IN A TEMPERATE ZONE DECIDUOUS FOREST ECOSYSTEM

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Abstract

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To assess atmospheric wet deposition impacts, a rainfall-runoff water quality monitoring study was performed in 2-forested experimental watersheds, in Turkey. Over a 2-year period some major ions were analyzed for 62 precipitation events while stream water was sampled at 15-day intervals in 2 subsequent water years; 1999–2000 and 2000–2001. A stochastic rainfall-runoff model was developed and used to estimate the discharge and then annual flux of the solutes to receiving streams. The pH in 83 percent of the precipitation events was lower than 5.6. The watershed's atmospheric deposition flux was in the higher ranks compared to other regions in the world. Because of the varying precipitation and deposition amount between the 2 years of monitoring, hydrogen ion deposition was 4.72 kmole ha^{-1} in the first monitoring year, while it was 1.70 kmol ha^{-1} in the second one. A net outflux was observed for Na^+ compared to significant accumulations of wet deposited sulfate, phosphate, nitrate, ammonium, potassium and magnesium in the watersheds.

Key words: acid deposition, deciduous forest, temperate zone, cation exchange

Introduction

The importance of atmospheric deposition as a nutrient source for forest ecosystems has been emphasized in many studies in the past 20 years (i.e. Haines, Swank, 1988; Likens, 1989; Hedin, Likens, 1996; De Hayes et al., 1999; Tanner, Wong, 2000). Furthermore, several long-term monitoring studies conducted in various regions of the world have shown that the deposition amounts vary significantly with time and some geographical attributes of the location including sea influence, altitude, climate and distance from pollution sources (McDonald, 1999; McCormick, 1998).

Haines and Swank (1988) have shown that acid rain could promote base cation leaching from the forest canopy, litter and soils. Hence, the chemical composition of runoff gives valuable information about retention, and biochemical transformations. Similar studies were carried out by Balcı et al. (1986) and Balcı et al. (1993) in Turkey.

Sulfur and nitrogen deposition studies are important contributions to the acid rain problem. Besides the studies related to nitrogen status and saturation in Europe and the USA (Swank, Vose, 1997; Knoepp, Swank, 1997; Aber et al., 1998; Boring et al., 1988, De Schijver et al., 2000), much researches has been directed to the effects of sulfur deposition (Johnson et al., 1980; Swank et al., 1984; Berner, Petsch, 1998; Fitzgerald et al., 1988; Johnson et al., 1999). Johnson et al. (1993) performed a simulation related to the effects of three S deposition scenarios – 50 % reduction, no change, and 100% increase – on the cycles of N, P, S, K, Ca and Mg in a mixed deciduous forest ecosystem in North Carolina, and found that increased S deposition caused substantial increases in Ca^{2+}, K^{+}, Mg^{2+} and SO_4^{2-} leaching over the 30 year simulation period. According to the results of another long term study in the same region covering 7 watersheds, more NO^{3-}N, NH^{4-}N and PO^{4-}P were added annually than are lost in streamflow for all watersheds. On the other hand, net losses occurred for Ca^{2+}, Na^{+}, K^{+}, Mg^{2+} and SiO_2 in all sites (Swank, Waide, 1998). Johnson et al. (1998) obtained similar results while Haines and Swank (1988) concluded that base cation leaching occurred at rainfall pH ≤ 5.5.

The synthesis of the above studies suggests that among many other processes, net losses of base cations could be an indicator of the extent of the acid deposition and the associated ecosystem response to it. In this study, atmospheric deposition to a temperate deciduous forest ecosystem has been investigated and its influences on some ecosystem processes has been evaluated on the watershed scale.

Material and methods

Site description

The study was conducted on two small sized watersheds located on the European side of Istanbul between Black Sea and Bosphorus (Fig. 1). The watersheds named as C and D are located adjacent to watersheds A (71.9 ha) and B (77.5 ha) which were previously studied by Balcı et al. (1986). A long-term paired watershed research project was conducted on watersheds A and B (Balcı et al., 1986), which helped interpolating the discharge values for C and D. Watershed C had an area of 150.3 ha with an elevation of 145.3 m (110–160) while D had an area and mean elevation of 134.5 ha and 122.3 m (100–145 m), respectively. The mean slopes of the watersheds were very similar. 21.6% for watershed C and 18.9% for watershed D.

The region is described as 'humid, oceanic and some water shortage in summer' according to the Thornthwaite method and classified as B3B1'sb4' (Serengil, Özhan, 2001). The nearest meteorological station (5 km) receives an average of 1050 mm of rainfall annually. Two different parent soil materials were determined in the region including carboniferous (clay grovak-schist) and neogen formations (upper pliosen) (Özhan, 1977). Study watersheds are located on neogen material that generated deep, acidic, coarse textured and graveled soils.

According to Yaltırık (1966) the region belongs to the Castanetum-Fagetum vegetation zone. The main tree species recorded in the watersheds were oak (*Quercus petrea*, *Q. robur* and *Q. frainetto*), beech (*Fagus orientalis*) and hornbeam (*Carpinus betulus*). The forests were mostly composed of the mixture or pure stands of these species having crown closures between 0.8–1.0 (1.0 represents the full closure of the crowns in a stand). Pure beech stands and beech dominated stands were generally located on the northern or eastern aspects while hornbeam preferred riparian zone. Many other shrubs and understorey species were observed in the watersheds dominated by *Rhododendron ponticum* L. and *Pteria equilina*.

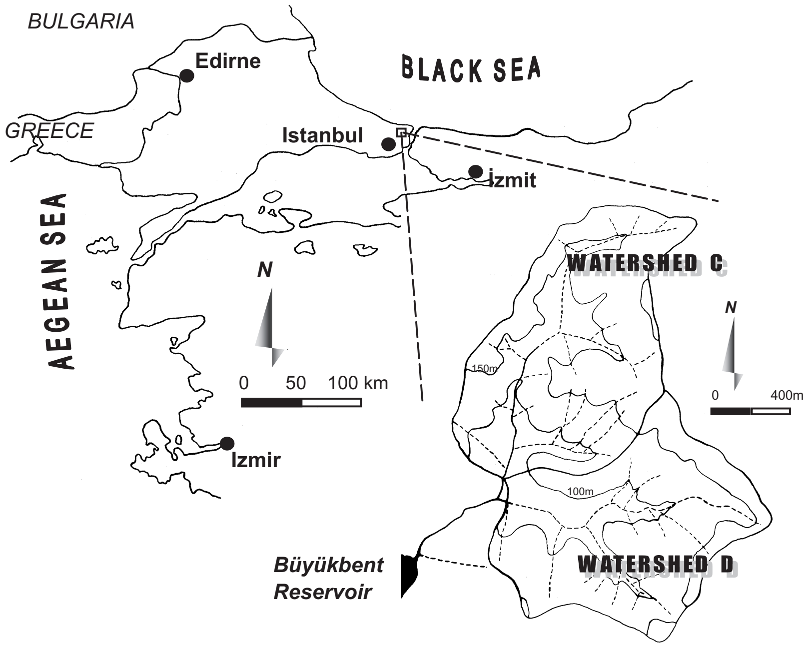

Fig. 1. Location of the study watersheds.

Sampling and analyses

The monitoring period was 2 hydrologic years (1999–2000 and 2000–2001). The hydrologic year starts in October and there is a dry period between July and September. Five sampling points (SP) were established on the streams (Fig. 2) and sampling was performed manually in 15 day intervals except for some extraordinary conditions (for instance, sampling was not done in case of no flow in dry summer season and was made more frequently during heavy rainfall events). The 3rd and 4th SPs were used as controls and to detect changes between 1st, 2nd and 5th SPs. The total number of stream water sampling occasions was 51. Rainfall was also sampled (total 62 events in 2 hydrologic years) and analysed for the same constituents as runoff which were pH, electrical conductance (EC), $Mg2^{+}$, K^{+}, Na^{+}, NO_3^{-}, SO_4^{2-}, NH_4^{+}, Al^{3+}, Fe^{2+}, Fe^{3+} and Cl^{-}. All water analyses were performed within 6 hours after collection. Only white plastic equipment was used and water samples were filtered with 0.45 μm membrane filter before the analyses (Deal et al., 1997). The precipitation water samples were taken from the samplers and analyzed immediately after the precipitation events.

An intensive systematic grid sampling pattern was prepared and implemented for soil sampling in both watersheds (Fig. 2). According to the previous studies performed in the region related to soil properties (Eruz, 1980) average A horizon depth was 0–20 cm, B horizon 20–60 cm and C horizon under 60 cm. Hence, soil samples were taken from these 3 depth levels. Some selected physical and chemical analysis were performed on these samples (texture, organic matter (OM) content, pH, EC, effective cation exchange capacity (ECEC) and base saturation (BS)) in order to assess soil buffering capacity.

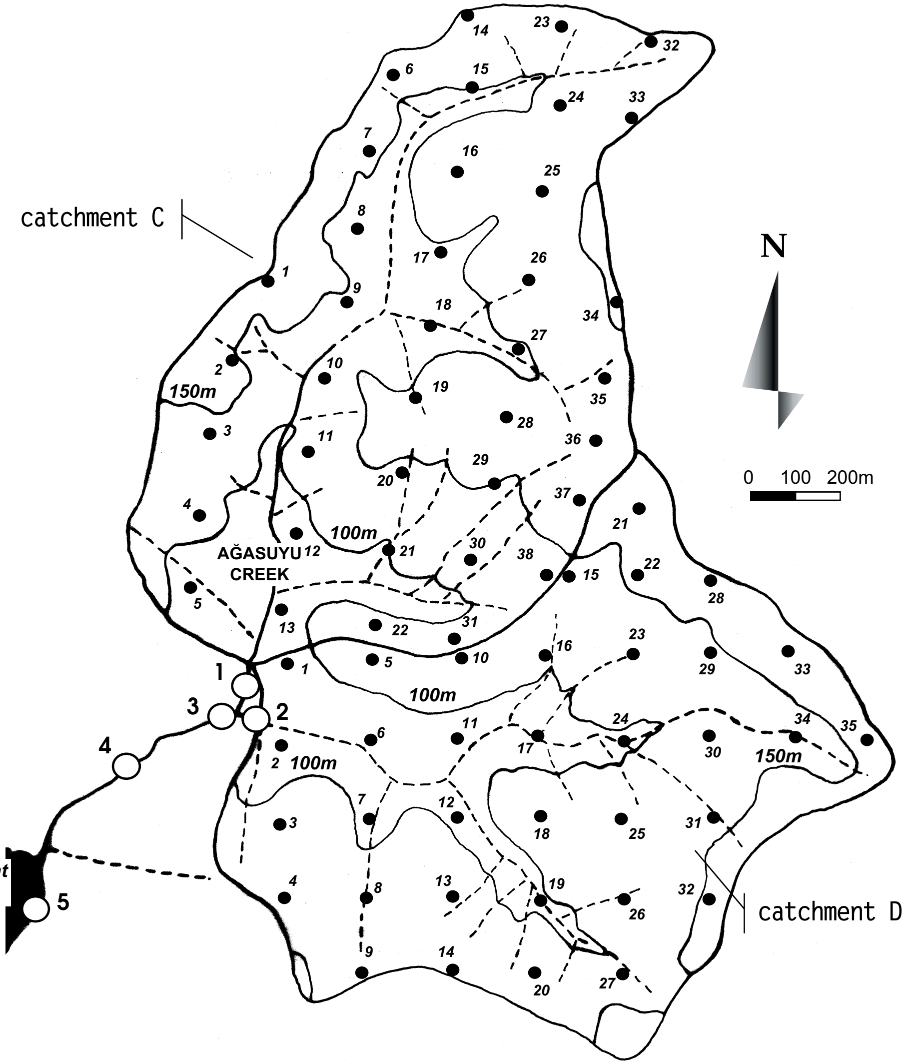

Fig. 2. Location of the water and soil sampling points (SP). Blank circles represent the water SP's (1-5), while dark ones represent soil SP's (1-38 in watershed C, 1-35 in watershed D).

Laboratory analytical methods

Nitrate, K^+, and Na^+ were measured using Orion ISE multimeter (ion selective electrode technique). Iron, aluminum, phosphate (all forms), magnesium, ammonium and sulfate were analyzed with DrLange digital photometer

(Ewing, 1975). Calcium and chloride were analyzed with titrimetric methods (APHA-AWWA-WPCF, 1975) and pH, dissolved oxygen (DO) concentration and (EC) with WTW Multiline pH meter.

Ammonium chloride was used as the solution for extracting cations from soil samples (Deal et al., 1997; Raben et al., 2000), and individually analyzed cations were added to calculate BS and ECEC (Sumner, Miller, 1996). OM percentage was determined with the Walkley-Black method while textures of the soil samples were determined with hydrometers (Irmak, 1956). Soil pH and EC measurements were performed on soil solutions prepared with distilled water. The ratio for pH and EC soil solutions were 1/2.5, and 1/5, respectively.

Data processing

All data were filtered according to the Quartile Estimation Method (Gürsakal, 1997) for quality assurance, and the statistical analyses were performed with SPSS 10.01 and MS Excell spreadsheet software.

Runoff estimation

In order to predict the outflow of the ions, streamflow must be measured or estimated. As stated above, streamflow for watershed C and D were interpolated from adjacent watersheds A and B. The procedure used in this study to interpolate the rainfall-runoff relationship is discussed below, briefly.

Interpolation model

As the watersheds in the region were all forested (conservation forest) and had similar vegetation cover, interpolation of streamflow within watersheds of similar size, relief and geology was thought to be a better solution instead of applying a totally new conceptual model. This statement is supported by the comparison of runoff coefficients calculated from 13 years of observed data from watersheds A and B (Fig. 3). The runoff coefficients of similar sized forest covered watersheds were close to each other as seen from the figure.

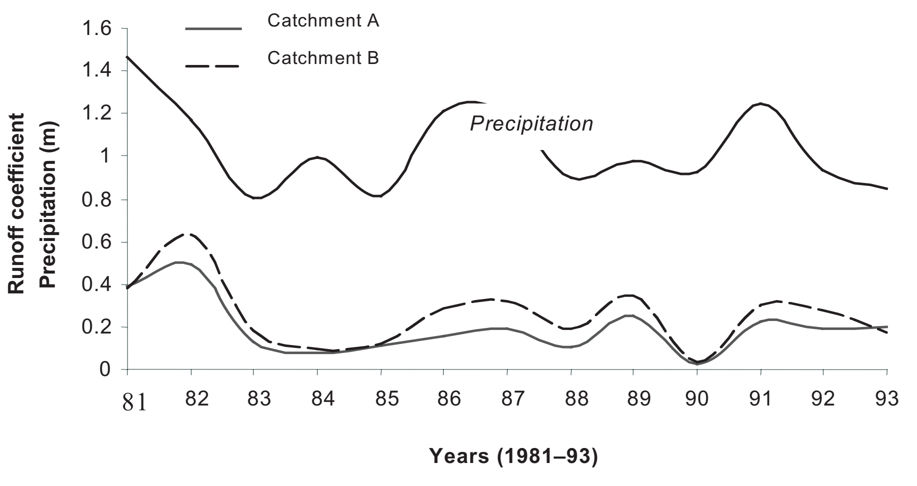

Fig. 3. Runoff coefficients of two experimental watersheds between years 1981 and 1993.

As a first step, rainfall-runoff characteristics of watershed A were investigated and a probabilistic model, based on nonlinear equations was constructed, tested for watershed B and then applied to C and D. In order to determine the history and the coefficients of the nonlinear equations, some statistical analyses explained below, were performed.

The history of a rainfall-runoff model can be determined with two methods; correlogram or trial and error (Müftüoğlu, 1984, 1991). The correlogram technique was used in this study. As the crosscorrelation coefficient became negative in the third year, the history was determined as the 3 subsequent years, including the current year. However because of the weak 2 lag crosscorrelation coefficient, effective history was accepted as 2 years (*including the current year*) for model construction.

This meant that the runoff during the current year was affected by the current years' and also the previous two years' precipitation, though the current and previous years' were thought to be most important. The same procedure was done for monthly data and a longer history was determined (*previous 3 months*). However only current and former months were taken into account to lessen the mathematical process.

Finally an empirical rainfall-runoff model (*given below*) was built for watershed A not only considering previous years or months precipitation but also the extreme rainfall amounts *(both for extreme months and years*). The extreme precipitation events (*more or less than average*) were adjusted with suitable mathematical models (*linear or nonlinear*) for each month.

Runoff of the nth month:

$Qn = EQ \pm \kappa$	EQ: a linear or nonlinear regression equation between current months rainfall and runoff,
	x_n : precipitation of the n^{th} month,
$\kappa = f\,(x_{n-1} - x_{n-1}mean)$	x_{n-1}mean: previous months average precipitation,
	x_{t-1}: t-1. Previous years precipitation,
Computed runoff for the year t	x mean: average annual precipitation.
	κ and γ: monthly and annual adjustment factors,
$Q_t = \sum Q_n \pm \gamma$	f (x): The best function to fit.
$\gamma = f\,(x_{t-1} - x\ mean)$	

Results

Hydrogen deposition and rainfall characterization

Rainfall of the region represents a significant amount of deposition of anions and cations (Table 1) considering the total precipitation of the hydrologic years 1999–2000 (1005.9 mm) and 2000–2001 (737.0 mm).

Elevation, distance to the sea, and pollution sources were thought to be the main factors responsible from the high deposition rate. Although the average pH value for the couple of years was 4.64, the distribution was slightly skewed to the left (Fig. 4) and thus median value is found to be 4.34 while the minimum and the maximum observed values were 3.30 and 6.95. The H^+ concentrations were approximately gamma distributed (*Shape* = 23.471, *scale* = 5.056).

The low pH values were detected mostly in the rainy winter months, particularly in January. On the other hand, in the dry summer months, the pH values were observed to increase over 5.6 (i.e. 6.25 for July, and 6.73 for August). When acidity of the precipitation was compared with wind direction, it was seen that the air masses coming from NW had relatively low pH values (pH = 4.31) compared to two other major wind directions NE (pH = 4.58) and SW (pH = 5.18) in the region. The difference was statistically significant

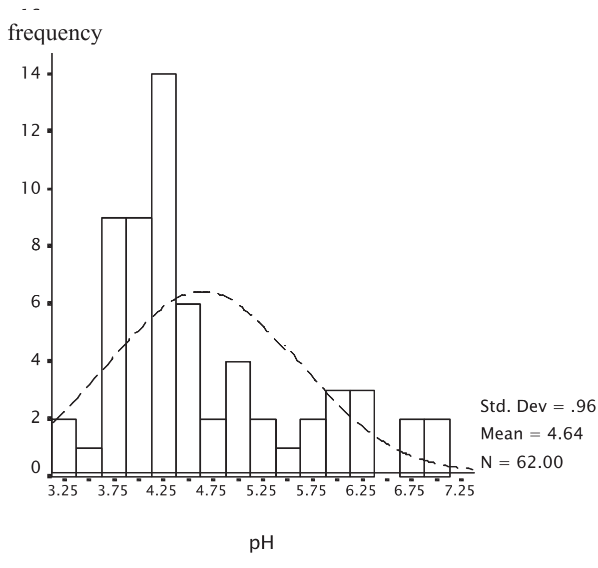

Fig. 4. Frequency distribution of precipitation pH values in the study site.

($p < 0.05$) and could be attributed to the acidic air masses mostly originating from Eastern European countries.

On the other hand, the equivalent concentrations of acid cations (23.21 μeq L^{-1}) were low compared to base cations. However, this is to be expected considering the predominance of anions, particularly sulfate and phosphate. Cations were dominated by magnesium while anions by sulfate.

T a b l e 1. Mean concentrations of the major ions in precipitation water.

Cation	Concentration (μeq l^{-1})	Anion	Concentration (μeq l^{-1})
H^+	22.82	$PO_{4-}^{2-,3-}$	74.53
Fe^{2+}	7.17	$SO4^{2-}$	108.22
Fe^{3+}	8.60	NO^{3-}	32.26
Mg^{2+}	70.78	Cl^-	24.64
Al^{3+}	0.39		
K^+	52.69		
Na^+	65.65		
NH^{4+}	19.37		
Total	**247.47**		**239.64**

Finally total wet deposition was calculated for the both years. In the first year (1999–2000) the annual precipitation was 1005.9 mm, resulting in a total deposition of 709.02 kmoles of hydrogen to the watershed C and 634.49 kmoles to watershed D (4.72 kmol ha^{-1} for both watersheds). In the second monitoring year (2000–2001) both precipitation (737 mm) and hydrogen deposition (1.70 kmol ha^{-1} for both watersheds) decreased. The amount of hydrogen deposition was 255 kmoles for watershed C and 228.81 kmoles for watershed D.

Soil analysis

The soil system represents one of the major components of a forest ecosystem. Furthermore it is the primary cation exchange subsystem. Many soil properties are affected by ion exchange which in turn affects the buffering capacity against acid deposition (Eruz, 1980).

The mean pH of the watershed soils decreased with depth (pH_{0-20}: 5.45; pH_{20-60}: 5.22; pH_{60-90}: 5.11 for watershed C and pH_{0-20}: 5.30; pH_{20-60}: 4.76; pH_{60-90}: 4.77 for watershed D). EC values of the soils in the reg ion were quite low and also decreased with depth (EC_{0-20}: 96.37 µS cm^{-1}; EC_{20-60}: 52.03 µS cm^{-1}; EC_{60-90}: 53.37 µS cm^{-1} for watershed C and EC_{0-20}: 142.11 µS cm^{-1}; EC_{20-60}: 54.51 µS cm^{-1}; EC_{60-90}: 83.06 µS cm^{-1} for watershed D). Conversely, mean clay percentage (CP) of the soils increased with depth (CP_{0-20}: 17.44%; CP_{20-60}: 33.72%; CP_{60-90}: 35.23% for watershed C and CP_{0-20}: 16.59%; CP_{20-60}: 27.69%; CP_{60-90}: 34.49% for watershed D).

Finally, organic matter (OM) contents of the watershed soils decreased with depth OM0-20: 11.36%; OM20-60: 5.56%; OM60-90: 5.54% for watershed C and OM0-20: 13.69%; OM20-60: 4.97%; OM60-90: 5.29% for watershed D) as expected. Due to suitable climatic conditions, the decomposition ratio is expected to be quite high in the region. Mull type litter had 5 to 15 cm thickness throughout the watersheds with a thin decaying sublayer underneath. The mean pH and electrical conductivity of this layer in both watersheds was 5.62 and 384.5 µS cm^{-1} respectively.

The CP was inversely proportional to OM percentage and increased with depth. This might have a balancing effect on the cation exchange behavior of the soil. Another increasing soil property with depth was hydrogen ion activity, which might stem from acidic parent material. The low electrical conductivity value and its decrease with depth is an indicator of the leaching and desorption of sodium and other salts from this ecosystem component.

Many chemical and physical soil properties, including organic matter content, amount and type of clay minerals and aluminum and iron oxides can influence the cation-anion retention and effective cation exchange capacities (ECEC) of soils (Pierzynski et al., 2000). Furthermore, the interaction of these soil properties with exchangeable cations might give us information related to the soil buffering system. Correlations among individually analyzed exchangeable cations and OM and CP were investigated on 60 soil samples from three depths.

CP had no correlation with exchangeable cations except aluminum ($r = 0.52$; $p < 0.001$) which also had linear relation ($r = -0.43$; $p < 0.001$) with OM content of soils. Exchangeable sodium and magnesium did not show a correlation with OM content and clay fraction while calcium, potassium, hydrogen, ECEC and BS correlated only with OM. Exchangeable cation amounts and effective cation exchange capacities of soils for soil depths are given in Table 2.

Base saturation, a significant indicator of the soils buffering system against acid deposition (Johnson, Todd, 1983) was very high for the soil samples taken from the study watersheds. Particularly, Ca^{2+} dominated the ECEC in the topsoil and the highest ECEC and BS values were determined in this horizon.

Table 2. Exchangeable cation content of soils at various depths.

Soil depth (cm)	N	Base cations				Acid cations		ECEC	BS
		Ca^{+2}	Mg^{+2}	K^{+}	Na^{+}	H^{+}	Al^{+3}		
		meq kg^{-1}	meq kg^{-1}	meq kg^{-1}	meq kg^{-1}	meq kg^{-1}	meq kg^{-1}	meq kg^{-1}	%
0-20	20	18.34	9.06	13.78	0.51	0.12	0.64	42.34	98.48
21-60	20	6.07	7.43	13.06	0.49	0.32	5.40	32.45	83.36
61-90	20	9.18	10.15	13.03	0.65	0.18	5.19	38.20	86.42

Note: N – number of samples.

Input-output relationships of important ions

Estimated input (*wet deposition*) and output amounts of some important ions are given in Table 3. Input values were computed by multiplying the monthly average concentration of the ion with the monthly precipitation (precipitation weighted). Output estimation was performed by the multiplication of the average ion concentration in stream water with the simulated monthly water yield (discharge weighed). Chloride is not placed in the table as the number of chloride analyses on the streamflow samples was not sufficient to make precise output estimation.

Table 3. Estimated input and output amounts of some important ions.

	Water years (October-June*)			
	1999–2000		2000–2001	
	Input	Output	Input	Output
	eq ha^{-1}	*eq ha^{-1}*	*eq ha^{-1}*	*eq ha^{-1}*
Al^{3+}	3.91	1.36	2.87	0.72
Fe^{2+}	72.11	45.15	52.83	23.77
Fe^{3+}	86.53	51.15	63.40	26.92
NH^{4+}	195.60	32.86	143.30	17.30
Mg^{2+}	**702.02**	**723.80**	514.35	380.96
K^{+}	806.53	184.81	590.93	97.27
Na^{+}	**660.40**	**1207.40**	**483.86**	**635.50**
NO_{3-}	244.43	120.26	179.09	63.30
$PO_4^{-,2-,3-}$	834.80	262.28	611.64	138.05
SO_4^{2-}	1494.77	421.17	1095.18	221.67

*July-September is the dry period. Annual runoff was 257.14 mm yr^{-1} in 1999–2000, 135.34 mm yr^{-1} in 2000–2001 water years. The calculation of equivalent phosphate was made for 3- charge.

According to the computed values above, the outflux of the nutrients in stream water rarely exceeded the amount of wet deposition. When dry deposition – not measured but considered to be in essential amount – of base cations were also taken into account, a net outflow could only be suggested for sodium. The measured anions were also retained in the forested watersheds. The outflux/influx ratio (O/I) of nitrate was slightly higher than phosphate and sulfate ($O/I_{NO3} = 0.42 > O/I_{PO4} = 0.27 > O/I$ SO4 = 0.24).

Influences on Büyükbent reservoir

Influence of atmospheric deposition on the runoff in forested watersheds is not clear enough owing to the complicated nature of such a system (Henderson et al., 1979). The input-output relations of the nutrients are known to be highly unpredictable, and variable. Furthermore, all ecosystems are unique with their nutrient cycling and filtering mechanism significantly varying with time. Despite the amount of atmospheric deposition, a very limited portion of the nutrients could reach the fluvial systems in the study region as stated above. This phenomenon thought to have 2 consequences which are:

- Atmospheric deposition of the ions could have very limited influences on stream water quality,
- The retention of some nutrients (i.e. N, P, K) could play a very significant role in increasing the life span of receiving reservoirs and lakes.

The seasonal variation of rainfall-runoff pH values might be an example to the first statement. The monthly mean pH value of precipitation water exceeded 6.5 only in August. However, the pH of the creeks never became less than 6.5 during the whole monitoring period (Fig. 5).

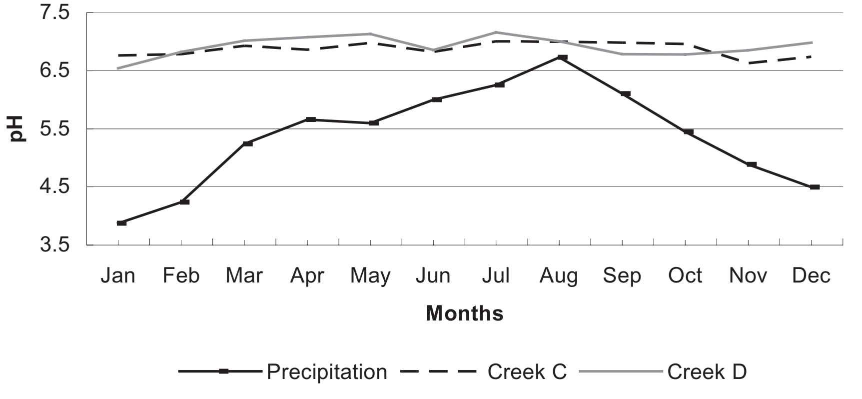

Fig. 5. Annual variations of pH values in stream and precipitation water.

The pH values of the creeks were quite stable, reflecting the regulating role of the forest ecosystem, while precipitation pH values had strong seasonal variations. As seen from Fig.

5, mostly winter months were subject to elevated acid deposition and even in this season, the forest ecosystem could continue its neutralizing function properly.

Natural or artificial lakes are known to have a certain life span influenced by many geographical and ecological features of the location. Nutrient supply is known as one of the key factors affecting this life span. Phosphorus and nitrogen are generally the dominating elements related to the nutrient enrichment of waterbodies and in turn promoting the autotrophic life in aquatic systems (Eugene, 1971).

The O/I ratio for nitrate, ammonium and phosphate were calculated as 0.42, 0.14, and 0.24 respectively, which means that less than half of the wet deposition of the mentioned ions were lost to the fluvial system. This filtering behaviour of the forest ecosystem acts a vital role in the region considering the state of the Büyükbent reservoir receiving water from the watersheds C and D. The reservoir was built 350 years ago and is currently filling with rushes and other aquatic plants. Lake water DO concentration was monitored throughout the study and it was found that the concentration became less than 5 mg l^{-1} during spring months.

Discussion

The deposition amount of hydrogen ion varied between the subsequent years significantly. More than 3 times more hydrogen ion deposited in the first monitoring year than the second year. Although the precipitation was acidic for most of the events throughout the year, base cation deposition was also in essential amount.

The dominant cation and anion in precipitation were magnesium and sulfate, respectively. Magnesium was followed in importance by sodium and potassium. Acid cations were low in equivalent concentration in precipitation water. Phosphate and nitrate also contributed significantly to total deposition. In spite of the relatively close proximity of the study site to the sea (*both horizontally and vertically*), chloride had the lowest concentration. Although watershed soils were not very efficient in cation exchange and acid buffering, they had high base saturation. Furthermore, the low outflux-to-influx ratio of anions from the catchments indicates the soils to be effective in anion retention, which also disables cation leaching. Particularly, sulfate retention and its subsequent biological transformation to the organic form might constitute an essential portion of the total sulfur pool in the forest soils (Swank et al., 1984).

Soil organic matter content was negatively correlated with acidic exchangeable cations (*hydrogen and aluminum*), whereas highly significant positive correlations were determined for exchangeable potassium and calcium. However no correlation was found between organic matter and exchangeable magnesium and sodium. Considering the high exchangeable calcium content in the topsoil we can suggest a strong relation between organic matter content and calcium and in turn cation exchange. Consequently, the upper soil horizon, is a major cation exchange layer and this horizon has more buffering potential than other subsoil horizons. This statement should be verified with more data and study for similar watersheds with acidic parent materials. If cation leaching from the soil is accepted as an indicator of the watershed response to acidic deposition, our results suggest that the amount of acid deposition measured in this study will not adversely affect the health of the studied ecosystems. However, any change in forest management practices (harvesting, thinning, changing species etc.) could cause essential alterations in nutrient cycles and thus affect the buffering system and cation pools of the watersheds.

Consequently, with the results of this study we can suggest the following statements:

- Acid precipitation is a reality for the lower latitudes, too.
- Substantial amounts of solutes other than hydrogen could deposit to the forest ecosystems.
- Acid neutralizing cations provided by decomposition and weathering process might be a key parameter to estimate buffering potential of the catchments.
- In the abundance of water and heat, the sufficient supply of phosphorus could promote the autotrophic life in the reservoir. However, the deciduous forest ecosystem in the region diminishes this effect, and in turn increases the life span of the reservoir.

Translated by the authors

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