VARIABILITY OF PHYSICO-CHEMICAL PARAMETERS IN PRECIPITATION IN POLAND (1996–1999)

BARBARA WALNA[1], ŻANETA POLKOWSKA[2], STANISŁAW MAŁEK[3], KRYSTYNA MĘDRZYCKA[2], JACEK NAMIEŚNIK[2], JERZY SIEPAK[4]

[1] Ecological Station of Adam Mickiewicz University, Jeziory, P.O.Box 40, 62-051 Mosina, Poland; e-mail: walna@amu.edu.pl (corresponding author)
[2] Chemical Faculty of Technical University of Gdańsk, Narutowicza 11/12, 80-952 Gdańsk, Poland
[3] Forest Faculty, Agricultural University of Cracow, 29 Listopada 46, 31-425 Kraków, Poland
[4] Laboratory of Water and Soil Analysis, Chemical Faculty of Adam Mickiewicz University, Drzymały 24, 60-613 Poznań, Poland

Abstract

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The paper presents the results of a research on precipitation carried out in the years 1996–1999 at three stations. They were then compared with those obtained at five other stations distributed throughout Poland, both in specially protected areas like national parks and those exposed to a strong detrimental impact of urbanised agglomerations and transborder pollution. The rainfall figures obtained differed markedly from those typical of the reference stations, while rain acidity has turned out to be considerable – its mean annual pH was often under 4.6. Electrical conductivity of precipitation in protected areas was in excess of 50 μS/cm, and indicating its pollution. Monthly patterns of pH and conductivity are given for both open terrain and throughfall, and tendencies of change are indicated. The ionic composition of precipitation was determined and sulphate concentrations were found to be decreasing with time. Sequences of anion and cation concentrations are given as well as the coefficients of throughfall modification.

Key words: precipitation, Poland, pH, ionic composition

Introduction

Poland, especially its southern and south-western regions, belonged to the most polluted areas in the 1980s (Grodzińska, 1990). Total SO_2 emission levels reached 4180 000 tonnes, nitrogen oxides, 1 550 000 t, and particulates 2 650 000 t. The area of convergence of the Polish, German and Czech boundaries known as The Black Triangle had the heaviest sulphur deposition (> 10 t/ha/y) in Europe (Nowicki, 1993). The latest figures (Environment, 2000) show a decline in SO_2 emissions in 1997 to 2 181 000 tonnes, nitrogen dioxide to 1 114 000 tonnes, and particulates to 1 130 000 tonnes. The year 1999 saw a further drop in the level of SO_2 to 1 897 000 tonnes and NO_x to 951 000 tonnes. Even more optimistic are the results of regional studies which show, for example, a sevenfold drop in the mean annual SO_2 concentration in the city of Poznań from 72 μg/m³ in 1987 to 11 μg/m³ in 2000 (Krysiak et al., 2000).

In Poland only some components of the environment have been observed to have improved. Monitoring-based studies of forest damage (Wawrzoniak, Małachowska, 2000) showed only a slight improvement in the health status of forests manifesting itself in an insignificant drop in the defoliation index (from 2.77 in 1998 to 2.70 in 1999). However, in terms of the percentage of trees not suffering defoliation, Poland closes the list of European countries with 10.4%; only Ukraine has worse figures (Seidling et al., 2001). The proportion of healthy trees (spruce, fir) is on the decline, pine shoots tend to die more and more often, and pathogenic fungi are an increasingly serious threat. These alarming developments can be partially explained by the fact that the acidity of precipitation is still very high in Poland (Polkowska et al., 1999; Walna, Siepak, 1999; Małek, Wężyk, 2000; Environment 2000; Małek, 2001; Walna, 2001; Mazurek, 2001).

This paper presents the results of a research on precipitation carried out at three stations and compares them with those obtained at five other stations located in both, specially protected areas (National Parks) and those exposed to high levels of pollution.

Study area

The research was conducted in three climatically different regions (Table 1, Fig. 1):

- Stara Piła, Rumia forest district, Pomeranian voivodeship (St. No. 1)

This area is highly diversified morphologically, which contributes to the wide differences in its climatic conditions. The chief feature of the climate is highly variable weather, both from day to day and from year to year. The region is situated in the temperate zone transitional between the oceanic and continental climates. Its climate is modified by the immediate neighbourhood of the Baltic. The sea and the coastal zone are a place where tropical air meets polar air. It has a direct impact on the pattern of climatic elements in the boundary layer of the atmosphere. The proximity of the sea makes winter mild in terms of temperature, lowers the temperature of summer, and maintains a high air humidity throughout the year. A year's list of weather forecasts shows the prevalence of winds from westerly and south-westerly quarters. The air circulation is longitudinal. Mean July temperatures do not exceed 18 °C, and in January they oscillate around 2–3 °C. Annual rainfall amounts to 550–600 mm (Polkowska et al., 1999).

- Wielkopolski National Park, Jeziory, Wielkopolska voivodeship (St. No. 2)

Wielkopolska voivodeship lies in the temperate zone, in an area where maritime and continental influences interpenetrate. This transitionality manifests itself mainly in the

T a b l e 1. Information about the research and reference stations analysing precipitation water in Poland.

Station No.	Reference	Sampling site	Geogr. location	Altitude a.s.l. [m]	Description of sampling area
1	This study	Stara Piła – Rumia	54°34' N 18°21' E	122	10 km south, large urbanised agglomeration of Gdynia and Gdańsk, near Baltic Sea
2		Jeziory	52°11' N 16°54' E	82	Wielkopolski National Park 15 km south of large urbanised agglomeration of Poznań
3		Chełmowa Góra –Ojców	50°12' N 19°49' E	430	Mountain area, Ojców National Park
4	Environment (2000), Air pollution in Poland (2001)	Łeba	54°45' N 17°32' E	2	Baltic coast
5	Mazurek (2001)	Borecka forest	54°09' N 22°04' E	157	Station with small pollution load
6	(Mazurek 2001)	Storkowo	53°47' N 16°30' E	90	Station with small pollution load
7	Environment (2000), Przybylska (2001)	Jarczew	51°49' N 21°59' E	184	Central Poland, lowland
8	Environment (2000), Przybylska (2001)	Mount Śnieżka	50°44' N 15°44' E	1.603	Sudety Mountains, heavy pollution from over southern and western borders

changeability of weather depending on the kind of inflowing air masses. Maritime polar air is also recorded in the region. It comes from the northern part of the Atlantic Ocean and is characterised by a relatively high water vapour content. In summer it flows in as a mass of cool air bringing high cloud amount and frequent rainfalls. In winter this air mass brings a warming and greater cloudiness. Continental polar air from E Europe or Asia is more rare here. The interaction of these two frequently alternating air masses is the chief determinant of the climate of this region. Prevailing winds are from the west, mainly westerly and south-westerly quarters. It is an area with the lowest rainfall in Poland, receiving an average of under 550 mm annually (Woś, 1994).

- Chełmowa Góra, Ojców National Park, Ojców, Małopolska voivodeship (St. No. 3)

The Ojców National Park is situated on the Cracow–Częstochowa Upland. The upland is a plate of Upper Jurassic limestone uplifted to about 300 m a. s. l. It inclines gently north-eastwards and is cut by tectonic faults on the south. Most winds blowing here come from the west. The diversified relief causes average rainfall to vary too. The heaviest rainfall is recorded on north-facing slopes and on tops (about 800 mm) while on valley floors situated in the so-called rain shadow it is lower (Małek, Wężyk, 2000).

Fig. 1. Distribution of the research and reference stations in Poland (the numbers of the authors' collection stations are on the black background).

Material and methods

Rainwater was sampled for four years at three stations in Poland. At all the stations bulk precipitation was collected in an open area and from under the trees. Collectors were located at 1.2 m above the ground. The location of the sampling stations is presented in Fig. 1 and a more detailed description of the places is given in Table 1. The table and the figure also present the location of the five reference stations: Łeba, No. 4, Storkowo, No. 5, Borecka Forest, No. 6, Jarczew, No. 7, and Mount Śnieżka, No. 8.

All the analyses were carried out within 96 hours of sample collection. To test them for the content of specific ions, use was made of equipment, which guaranteed reliable and reproducible results. Measurements of pH were obtained using a CX 315 microcomputer pH-meter, while those of conductivity, a RADELKIS OK-102/1 analogue conductivity meter. Chemical analyses were performed after filtering (0.45μm). Concentrations of K^+ and Na^+ ions were determined using a Carl Zeiss Jena FLAPHO 4 flame spectrophotometer, and those of Mg^{2+} and Ca^{2+} ions with the help of a BUCK Scientific MODEL 210 VGP or Varian 20 atomic absorption spectrophotometer, for anions use was made of ion chromatography (Dionex-100, Dionex-320, Dionex-500), and for ammonium the colorimetric technique using MERCK SQ-118 spectrophotometer. A detailed description of the experimental methods, their calibration and validation based on a certified reference material (Major Elements in Rain Water No. 409, Commission of the European Communities, Community Bureau of Reference – BCR) can be found in the following references: Polkowska et al., 1999; Walna, Siepak, 1999; Małek, 2001. The amount of the rainfall was measured using Hellman's collector.

Results and discussion

Rainfall

Out of the three research stations, the heaviest rainfall was recorded at station No. 1 in 1998 (914 mm) and 1999 (869 mm). In the latter year, at station No. 3 the smallest volume was recorded (394 mm). At the remaining stations rainfall figures varied with the location. Maximum volumes were observed at the mountain station (No. 8) in 1998 and 1999 – over 1200 mm, while minimum ones were recorded at all the other stations in 1996: station No. 5 – 510 mm, station No. 6 – nearly 700 mm and station No. 7 – 564 mm (Fig. 2).

Rainfall figures at the research stations differ with the season. All of them have a summer peak, while in winter, or sometimes in spring, rainfalls are lower. Accordingly, the highest rainfall volumes were recorded at stations No. 1 and 3 in 1997 in summer (No. 3 – 354 mm) and autumn (No. 1 – 356 mm), whereas the lowest figure was recorded in the winter of 1995/96 at station No. 2 (43 mm).

Precipitation pH

The basic measure of pollution of precipitation is its pH. Fig. 3 shows a map with annual pH values for the research and reference stations. The least acidic rainfall (pH = 5.15) was recorded in 1996 at station No. 1, at which precipitation generally has the highest pH values out of all the eight stations. The most acidic sample (pH = 3.90) was collected in 1996 at station No. 3, which gets the most acidic rainfall with the pH oscillating around 4.00. Over

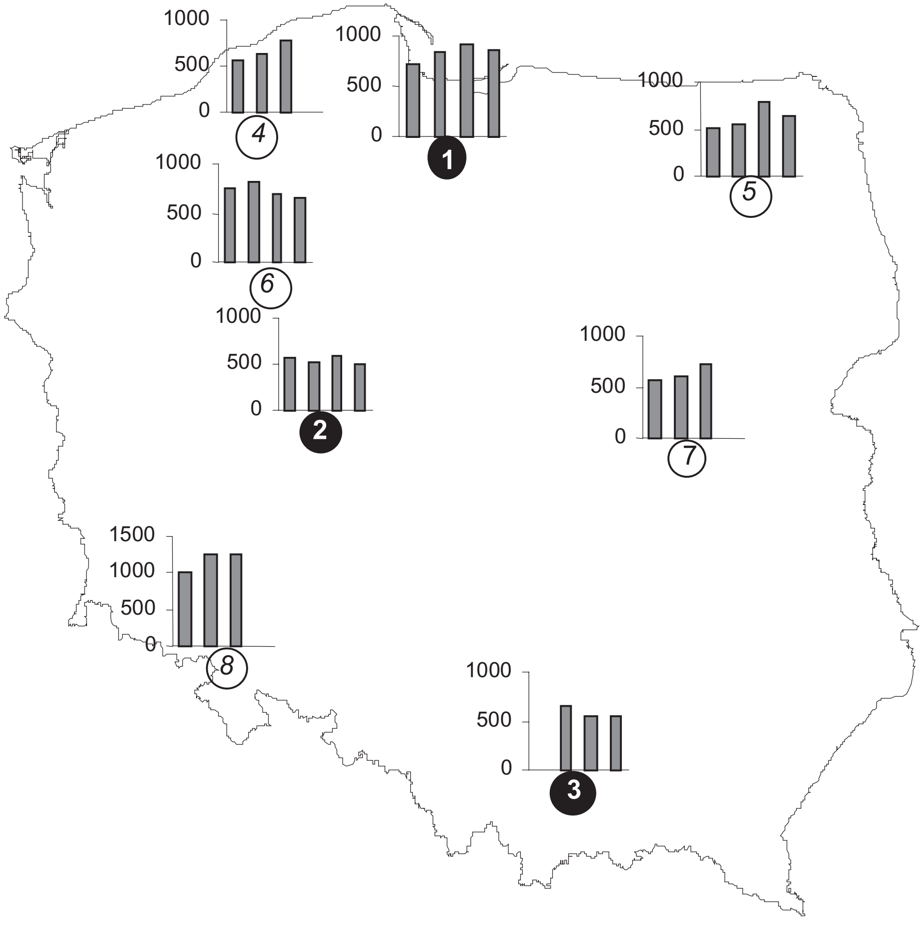


Fig. 2. Annual rainfall (mm). Successive bars in the diagram correspond to the years 1996, 1997, 1998, and 1999 (the numbers of the authors' collection stations are on the black background).

the four study years, at station No. 2 a slight upward tendency was observed, from pH 4.15 in 1996 to pH 4.52 in 1999. The pH of rainfall at the reference stations is also low, both at No. 8 situated near the area which has the lowest rain pH in Europe and at No. 5, located in a region free from pollution sources where the rain pH exceeds 4.5 only slightly.

The pH values of monthly rainfall for station No. 1 are much higher than for the remaining ones. Here the maximum values were even in excess of 7.0 (7.23 in October 1999) and a few times, over 6.0. The lowest figure for this station was 3.89 (July 1998). The rain pH in consecutive months can vary considerably. Over the four years a downward tendency

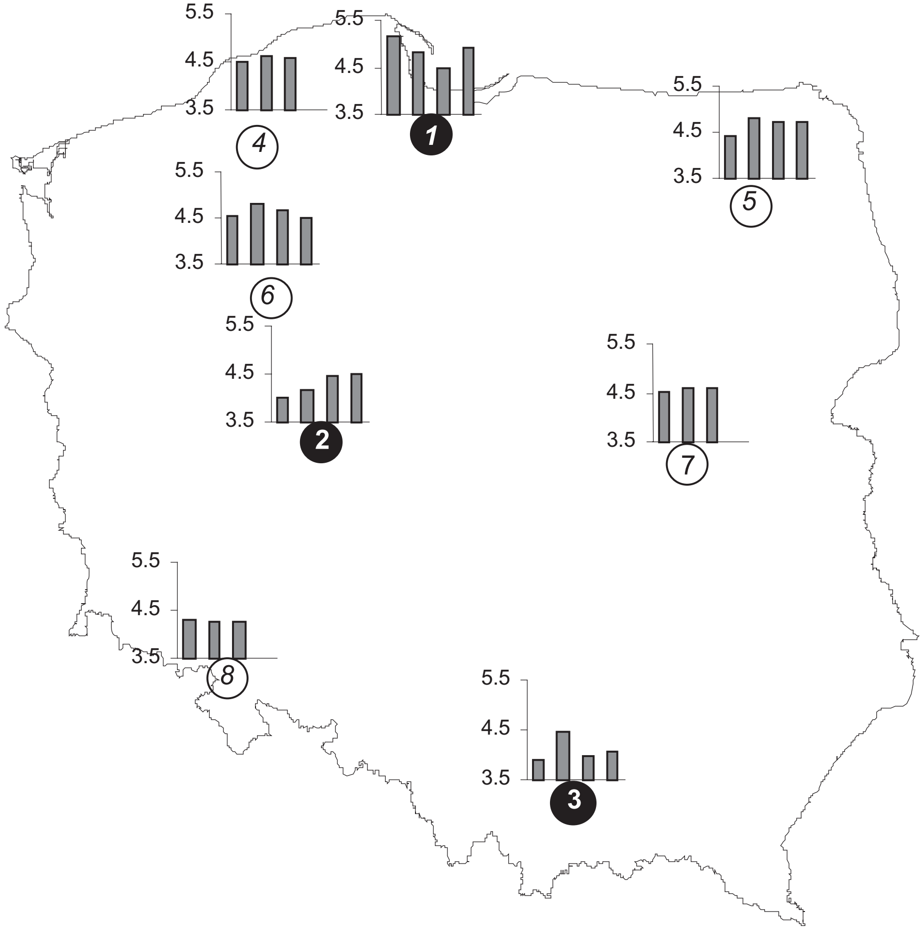


Fig. 3. Mean annual pH values at the research and reference stations. Successive bars in the diagram correspond to the years 1996, 1997, 1998, and 1999 (the numbers of the authors' collection stations are on the black background).

was observed. At stations No. 2 and 3 the pH variation patterns were similar and showed a slight upward tendency. The lowest monthly pH means recorded were 3.49 (station No. 2, January 1997) and 3.30 (station No. 3, March 1999).

It is interesting to compare the physico-chemical properties of rainfall collected in the open with that from under tree crowns. At the research stations precipitation was collected under spruces (station No. 1), oaks (station No. 2), and beeches (station No. 3). Fig. 4 presents mean annual pH values for the three research stations and a reference one (station No. 6, spruce). In 1996 the modification of rainfall after it had passed through spruce crowns

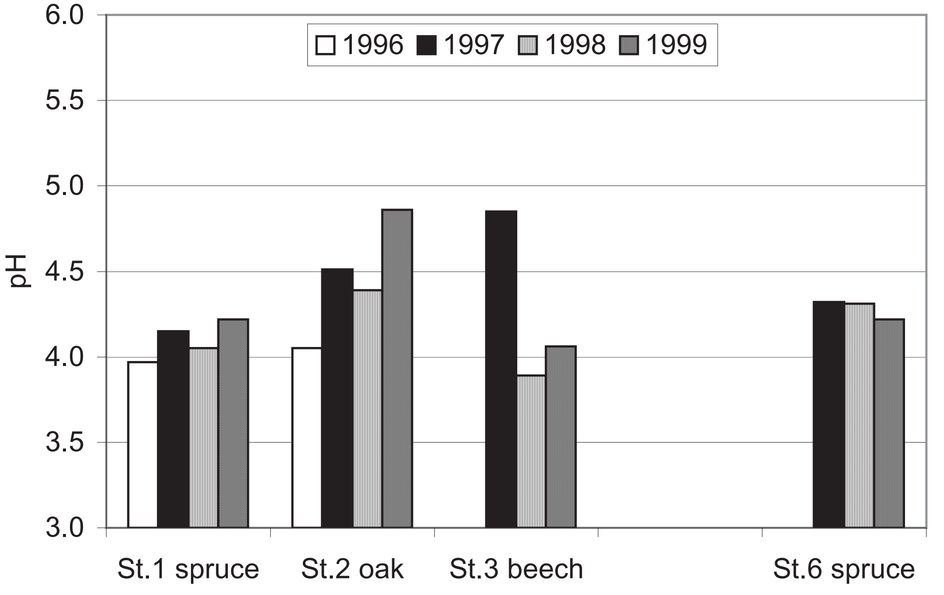


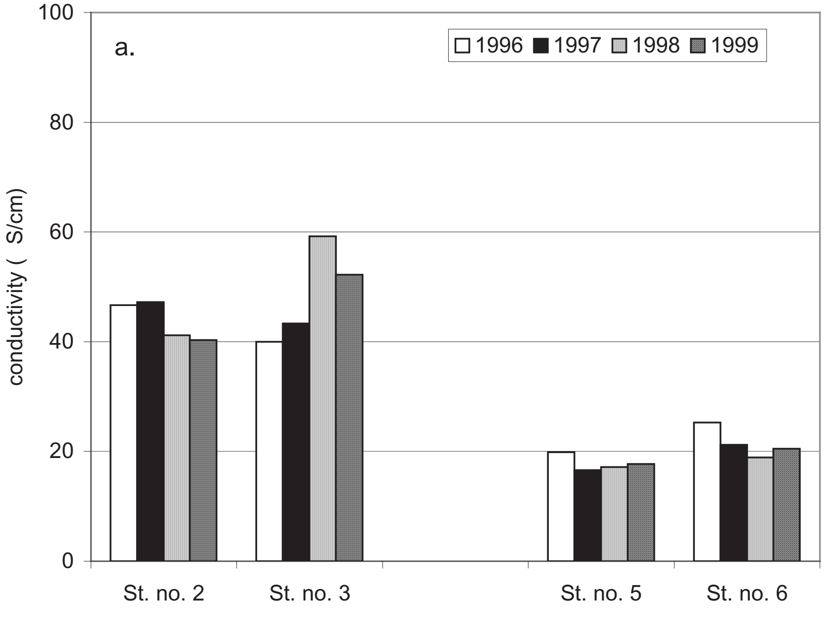
Fig. 4. Mean annual pH values of throughfall at the three research stations (No. 1, 2, 3) and a reference station (No. 6).

(No. 1) caused its pH to drop from 5.15 (open area) to 3.97 (throughfall). In the other years the differences were 0.6 (1997), 0.4 (1998), and 0.7 (1999). The influence of the oak crown (station No. 2) was only slight and tended to increase the pH, e.g. in 1997 from 4.18 to 4.51. The beech crown (station No. 3) had a similarly slight effect on the pH.

Precipitation conductivity

Electrical conductivity is a parameter well characterising the amount of dissolved and dissociated ions in precipitation. The highest annual values of electrical conductivity was observed at station No. 3 in 1998 – 59µS/cm (Fig. 5a). The lowest value was recorded at station No. 1 in 1996 – 21 µS/cm (it is not shown in the diagram because of the one-year measuring cycle). The conductivity at reference stations No. 5 and 6, situated far from pollution sources, was much lower – about 20 µS/cm.

Monthly conductivity showed a very high values recorded at station No. 2 in January 1997 – 225 µS/cm, and at station No. 3 in February 1998 – 188 µS/cm and in March 1999 – 170 µS/cm. The lowest was observed at station No. 1 – 7 µS/cm (November 1996) and 9 µS/cm (July 1996).



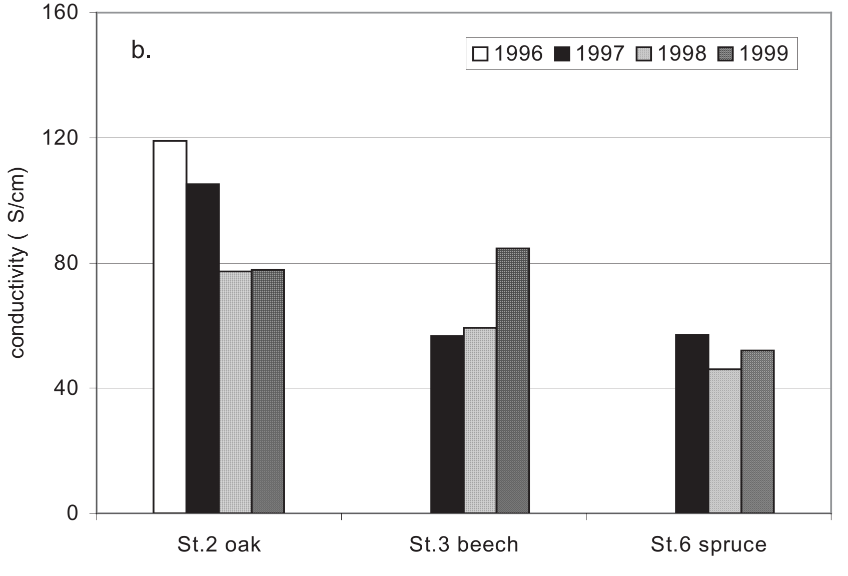


Fig. 5a. Mean annual values of rainfall conductivity at research stations No. 2 and 3 and reference stations No. 5 and 6 (open area).
Fig. 5b. Mean annual values of throughfall conductivity at stations No. 2, 3 and 6.

Like the acidity of rain, its conductivity is also modified significantly by the tree canopy (Fig. 5b). The lowest mean annual conductivity values were recorded at station No. 1 (not included in Fig. 5b) in 1996 – 49 µS/cm, and No. 3 in 1997 and 1998 – 57 and 59 µS/cm, respectively. The highest was observed at station No. 2 in 1996 – 119 µS/cm. Throughfall conductivity at station No. 2 was much higher (95 µS/cm) than at No. 3 and 6 (67 and 52 µS/cm, respectively). The employed index of modification of the rainfall composition after it had passed through tree crowns was Moore's (1987) enrichment coefficient, which is a conductivity/concentration ratio in precipitation under trees and in the open (Table 2). Enrichment coefficients were the lowest at station No. 3 under beeches, and the highest, at reference station No. 6 under spruces.

Maximum values of monthly throughfall conductivity were recorded at station No. 2 (326 µS/cm in May 1996, 401 µS/cm in January 1997, and 290 µS/cm in November 1997), and minimum, at stations No. 1 and 3 (about 20–30 µS/cm).

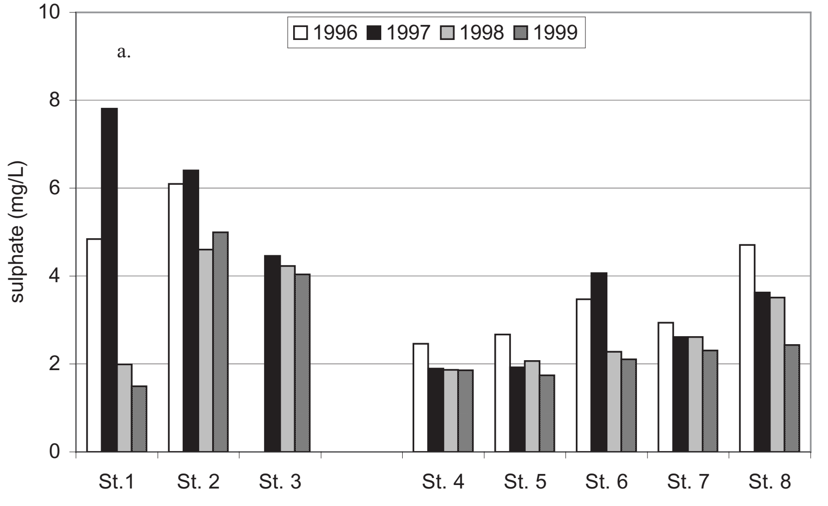
Table 2. Enrichment coefficients (conc. in throughfall / conc. in open) of individual ions and conductivity at the three research stations in the years 1996–1999.

Station No.	Year	Cl^-	NO_3^-	SO_4^{2-}	PO_4^{3-}	Na^+	K^+	Mg^{2+}	Ca^{2+}	NH_4^+	Conductivity	
1	1996	3.6	2.0	3.9	3.5	3.1	6.9	3.5	1.4	2.4	2.3	spruce
	1997	5.7	4.7	3.1	2.8	2.9				4.5		
	1998	5.3	3.8	7.6	3.9	3.1				2.4		
	1999	4.2	2.4	7.6	2.6	2.7				1.7		
2	1996	1.1	2.9	1.1	2.0	3.6	2.1	6.3	1.8		2.5	oakoak
	1997	1.2	1.7	2.3		3.5	7.3	5.9	2.0		2.2	
	1998	1.1	1.4	3.5		1.9	5.1	4.5	2.1		1.9	
	1999	1.2	1.3	1.2	2.5	3.8	1.8	5.5	1.2		1.9	
3	1997	3.6	2.9	2.3		1.8	2.4	4.5	3.9		1.3	beech
	1998	1.3	3.9	1.5			2.4	1.5	2.5		1.2	
	1999	9.7	5.1	2.6		1.4	2.6	3.8	3.2		1.6	

Chemical composition of precipitation

Over the study period there was a marked decline in the concentrations of sulphate ions at all the stations. The change was the most conspicuous at station No. 1, from about 8 mg/l in 1997 to 1.5 mg/l in 1999. A smaller decrease was observed at stations No. 2 (from about 6.3 mg/l to 4.8 mg/l) and 3 (4.5 mg/l to 4.0 mg/l). A downward tendency was also noted at stations No. 4, 5, 6, 7, and 8. Extreme annual concentrations of sulphate ions at those stations did not exceed 5 mg/l (Fig. 6a).

The highest annual nitrate concentrations were observed at station No. 1 in 1996 – 10.2 mg/l. This was due to the very high concentrations recorded in December – 24.3 mg/l, and from May to August – about 13 mg/l. The concentrations were the lowest at station No. 2, from 0.6 to 1.6 mg/l, while at No. 3 nitrate concentrations increased in 1999 to 3.2 mg/l. At stations No. 4, 5, 6, and 7 the concentrations remained at a steady 2 mg/l (Fig. 6b).



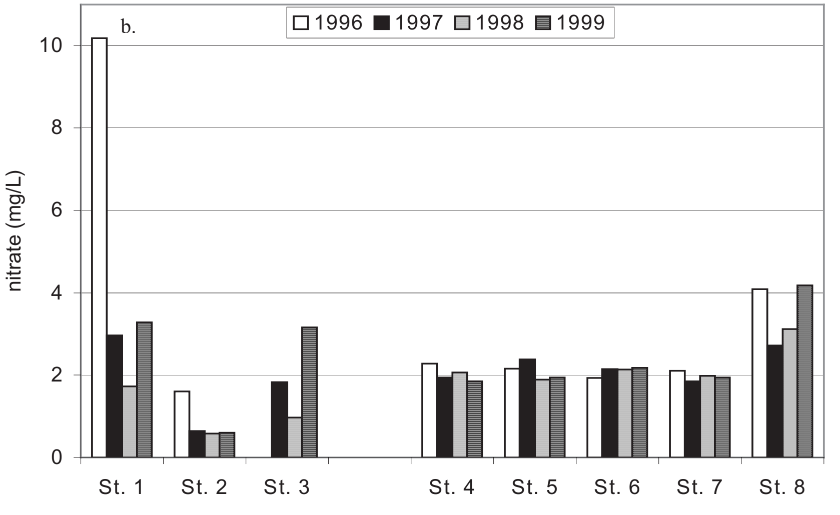


Fig. 6a. Mean annual concentrations of sulphate ions at the research stations (No. 1, 2 and 3) and reference stations (No. 4, 5, 6, 7 and 8).
Fig. 6b. Mean annual concentrations of nitrate ions at the research stations (No. 1, 2 and 3) and reference stations (No. 4, 5, 6, 7 and 8).

In the composition of precipitation in open terrain at station No. 1, the following sequence of ions was observed:
Ca^{2+} (2.5 mg/l) > NH_4^+ (1.0 mg/l) > K^+ = Na^+ (0.4 mg/l) > Mg^{2+} (0.2 mg/l).

Throughfall collected under the spruce showed a bit different sequence:
Ca^{2+} (3.4 mg/l) > K^+ (3.0 mg/l) > NH_4^+ (2.5 mg/l) > Na^+ (1.3 mg/l) > Mg^{2+} (0.7 mg/l).

Anion concentrations in precipitation showed the following pattern:
NO_3^- (4.6 mg/l) > SO_4^{2-} (4.0 mg/l) > Cl^- (1.4 mg/l) > PO_4^{3-} (0.15 mg/l).

A different regularity was observed in throughfall from under the spruce:
SO_4^{2-} (17.3 mg/l) > NO_3^- (12.3 mg/l) > Cl^- (6.6 mg/l) > PO_4^{3-} (0.4 mg/l).

For station No. 2 the respective sequences looked as follows (open terrain):
Ca^{2+} (1.4 mg/l) > K^+ (0.8 mg/l) > Na^+ (0.7 mg/l) > Mg^{2+} (0.3 mg/l);
under the oak:
Ca^{2+} (2.6 mg/l) > K^+ (2.4 mg/l) > Na^+ (2.0 mg/l) > Mg^{2+} (1.5 mg/l).

Anions (open terrain):
SO_4^{2-} (5.5 mg/l) > Cl^- (3.4 mg/l) > NO_3^- (0.86 mg/l) > PO_4^{3-} (0.15 mg/l)
under the oak:
SO_4^{2-} (10.9 mg/l) > Cl^- (3.9 mg/l) > NO_3^- (1.8 mg/l) > PO_4^{3-} (0.35 mg/l).

The precipitation composition in the open at station No. 3 displayed yet another ion sequence: K^+ (1.6 mg/l) > Ca^{2+} (1.3 mg/l) > Na^+ (0.4 mg/l) > Mg^{2+} (0.2 mg/l);
under beeches: Ca^{2+} = K^+ (4.0 mg/l) > Na^+ (0.8 mg/l) > Mg^{2+} (0.7 mg/l).
For anions in open terrain: SO_4^{2-} (4.2 mg/l) > NO_3^- (2.0 mg/l) > Cl^- (0.7 mg/l)
under beeches: SO_4^{2-} (9.1 mg/l) > NO_3^- (8.4 mg/l) > Cl^- (2.0 mg/l).

Table 2 lists enrichment coefficients for individual ions in throughfall at the three research stations in the years 1996–1999. They range from 1.1 to 9.7 for anions and from 1.2 to 6.9 for cations.

Conclusion

For four years (from 1 January 1996 to 31 December 1999) precipitation samples were collected at three research stations (Rumia, Jeziory and Ojców). The samples were then tested for pH, conductivity, and the amount of rainfall, as well as for selected anions and cations. The obtained sets of results were processed to extract the fullest information possible concerning both ion concentrations and interrelations among the physico-chemical parameters describing the precipitation water collected in three different woodland areas. The measuring data obtained during the project as well as data from literature provide a basis for the following conclusions:

- At all the research and reference stations the mean annual pH was lower than 5.1, and very often dropped below 4.6 (very acidic). This situation, especially at Ojców, can be explained by a direct impact of the industrial pollution load the areas receive due disadvantageous directions of the prevailing winds. The two national parks are among the most polluted protected areas in Poland.

- One can observe that at six out of the eight stations pH values were the lowest in 1996. Only at the Śnieżka station (No. 8), which had very low pH figures throughout the study period anyway, precipitation acidity remained fairly constant at pH 4.2–4.3.
- The pH was found to be greatly reduced in spruce throughfall in comparison with open terrain. At Jeziory (No. 2, oak) and Ojców (No. 3, beech) there was no distinct change, or the pH of throughfall was slightly increased. At all the three stations the pH of precipitation collected under the trees showed an upward tendency.
- The electrical conductivity of precipitation sampled at Jeziory (No. 2) and Ojców (No. 3) ranged between 40 and 59 µS/cm, which qualifies them for the group with 'high' and 'increased conductivity'. The rainfall collected at Rumia (No. 1) as well as in the Borecka forest (No. 5) and Storkowo (No. 6) had much lower conductivity (17–25 µS/cm), which allows them to be classified as ones with 'slightly increased conductivity'.
- At all the research and reference stations a distinct decline in the concentrations of sulphate ions was recorded (74% at Rumia, No. 1, 22% at Jeziory, No. 2, 49% at Storkowo, No. 6, and 26% on Mount Śnieżka, No. 8).
- The sequences of cations and anions in the ionic composition of throughfall were similar to those recorded in the open, while the concentrations of individual ions increased depending on the species of trees growing in the area, their age, and the acidity of precipitation.

Translated by M. Kawińska

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