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VARIABILITY OF THROUGHFALL AND STEMFLOW DEPOSITION IN PINE AND BEECH STANDS (CZARNE LAKE CATCHMENT, GARDNO LAKE CATCHMENT ON WOLIN ISLAND)

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Abstract: The research sought to determine the range of conversion of the chemical composition of precipitation in a beech stand located in the Gardno Lake catchment (Wolin Island) and a pine stand in the Czarne Lake catchment (Upper Parsęta catchment, West Pomerania Province). The presented results cover three hydrological years: 2012, 2013 and 2014. The research focused on the chemical composition of bulk precipitation (in the open), throughfall and stemflow (in a forest). The obtained results confirm that after precipitation has had contact with plant surfaces there is an increase in its mineral content. This is due to the process of enrichment of throughfall and stemflow with elements leached out of needles and leaves (K^+, Mg^{2+}) and coming from dry deposition (NH_4^+, Cl^-, Na^+, Mg^{2+}, SO_4^{2-}, NO_3^-) washed out from plant surfaces. The research, based on the canopy budget model, indicates that in the case of potassium, its load leached out of needles and leaves accounted for 75.6% and 73%, respectively, of its total deposition on the forest floor. Calcium leaching was not detected in either of the two stands. In comparison with potassium, the range of magnesium leaching was smaller and amounted to 34% under beech and 26.5% under pine. As to loads of potassium and magnesium in the beech stand, they were more than twice as large as the ones observed in the pine stand. In spite of the fact that coniferous trees capture aerosols present in the air much more effectively, a higher mineral content was recorded in the beech stand. It applies primarily to ions of marine origin (Cl^-, SO_4^{2-}, Na^+, Mg^{2+}). It was only when ammonium ions originated from agriculture that their higher concentrations and loads were found in the pine stand.

Keywords: forest ecosystem, throughfall, stemflow, ion deposition, canopy budget model, Scots pine, European beech

Introduction

Precipitation flowing through the canopy in the form of throughfall (TF) and stemflow (SF) substantially alters its physico-chemical properties when in contact with plant surfaces. The main factors which affect the course, character and intensity of these processes include: an atmospheric supply of various ions and their concentration levels in leaves, the species composition and age of a stand, and the abundance of nutrients in a habitat (Stachurski 1987; Swank 1986). These processes include the enrichment of precipitation through washing off dust and gas pollutants deposited on plant surfaces as well as through leaching elements from needles and leaves (van der Mass, Pape 1991; Likens, Borman 1995; Grodzińska, Laskowski 1996; Dambrine *et al.* 1997; Draaijers *et al.* 1997; Walna, Siepak 1999; Polkowska *et al.* 2005; Małek *et al.* 2008; Kozłowski *et al.* 2012). Consequently, despite much lower precipitation totals in the forest (interception), its soil is provided with a much larger flux of mineral and organic components than an open area. It is assumed that coniferous trees modify the chemical properties of precipitation much more than deciduous ones (Shubzda *et al.* 1995; Fernandez-Sanjurjo *et al.* 1997; Herrmann *et al.* 2006; Kozłowski *et al.* 2012). Having needles throughout the year and a larger reception surface (compared with leaves), they contribute to increased supplies of pollutants to the soil.

The processes taking place in the canopy affect the acid-alkaline balance of precipitation in woodland. Airborne nitrogen and sulphur compounds can be adsorbed on plant surfaces and acidify throughfall and stemflow (Gower *et al.* 1995; Walna, Siepak 1999; Kvaalen *et al.* 2002; Bochenek *et al.* 2008). On the other hand, in the case of deciduous trees, the canopy neutralises acidic compounds. As a result, the pH of throughfall is higher than of precipitation in the open.

The primary objective of the research was to:
- get familiar with the process of modification of the chemical composition of precipitation in tree stands of different species composition,
- determine the total canopy leaching of Ca^{2+}, Mg^{2+} and K^{+},
- identify changes which take place in the tree canopy in terms of precipitation acidity.

Study area

The research sites were located in the Czarne Lake catchment in West Pomerania (the upper Parsęta catchment), 8 km north-west of Szczecinek (the pine stand) and on the Wolin Island in the Gardno Lake catchment (the beech stand) (Fig. 1). The upper Parsęta catchment is situated on the border between two mesoregions:

the Drawskie Lakeland and the Bytowskie Lakeland (Kondracki 2013). The area of the Czarne Lake catchment is 17.66 ha, 3.1 ha of which is the lake itself. The catchment is thoroughly covered with woodland, mostly composed of clusters of fresh pine trees. The research was conducted in a pine stand situated in the Szczecinek Forest Division, the Szczecinek forest district, division 36d. This stand is 95 years old and represents a suboceanic *Leucobryo-Pinetum* fresh pine type of forest. The pine is the dominant tree species (85.8%) with an estimated canopy density of 70%. Its average DBH (diameter at breast height) is 33.8 cm and the average height is 25 m. Apart from pines, there are also silver birches (*Betula pendula*) – 2.3%, European beeches (*Fagus sylvatica*) – 4.3%, and common oaks (*Quercus robur*) – 6.6%.

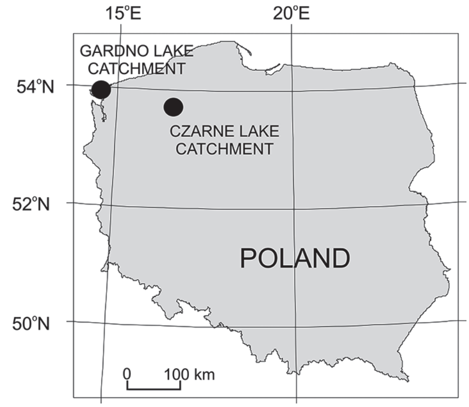

Fig. 1. Location of the research sites
Source: authors' own study.

The soil cover at the first research site was classified as a subtype of podzolic rustic soil formed from medium-grained sand sediments. The other research site was in the Gardno Lake catchment, 2.65 km^2 in area, situated in the Uznam and the Wolin mezo-region. The individual character of the catchment derives from its coastal location, proximity to the cliff coast and its full afforestation. The site was located in a forest ecosystem, in a poor habitat of acidophilic beeches referred to as a *Luzulo-pilosae-Fagetum* association. Predominant in its species composition was the European beech (*Fagus sylvatica*), at 90%, estimated to be 113 years old. The rest was composed of the Scots pine (*Pinus sylvestris*) aged 103. Their average DBH (diameter at breast height) was 35 cm and 42 cm, respectively, and the average height was 26 m for both species. The canopy density was found to be full. Beeches grow on sandy soils composed of medium-grained loose (rarely weakly loamy) sands. They form sandy luvisols with some glossic features, podzolised, with a very acidic pH, especially in the upper part of the soil profile.

During the study period, the total annual atmospheric precipitation and the mean annual air temperature in the upper Parsęta catchment amounted to 698.3 mm and 8.1°C (2012), 582.2 mm and 7.5°C (2013), and 539.9 mm and 9.2°C (2014). According to a thermal-precipitation classification (Lorenc 1999), the period can be regarded in thermal terms as average (the years 2012 and 2013) and very warm (2014). In terms of annual precipitation totals, the year 2012 was average, 2013 dry, and 2014 very dry. These values were obtained at the Research Station in Storkowo located at a distance of 5 km from the Czarne Lake catchment. As for the other catchment, the total annual precipitation and average annual temperatures were: 575.5 mm and 9.3°C (2012), 629.5 mm and 8.8°C (2013), and 592.9 mm and 10.4°C (2014) (the Biała Góra Research Station). Against the background of multi-annual figures (the years 1956–2014, Dziwnów) the period of the hydrological years 2012–2014 was relatively warm in thermal terms and average in precipitation terms.

Research methods

The presented results cover a period of three hydrological years: 2012, 2013 and 2014. The measuring system in the pine tree stand consisted of 12 throughfall collectors located under the tree canopy at nodes of a regular grid, distanced every 4 metres (Photo 1). Collector inlets were placed 20 cm above the ground and secured with a mesh to prevent contamination with organic material. The total inlet area of the collectors was 1.356 cm^2. The funnels and receptacles were made of plastic which did not affect the chemical composition of precipitation. In order to reduce any water losses through evaporation, the containers were buried in the ground with PVC pipes placed in the middle. In the beech stand the measuring system consisted of 10 collectors arranged in a grid. Every collector inlet was placed at a height of 1 m above the ground. The total inlet area of the collectors was 2.000 cm^2. The differences in the organisation of the measuring system relate to those in the structure of the tree stands examined (their canopy density, vertical structure, species composition).

In the beech stand stemflow was monitored as well. Three beeches located in the study area were selected for this purpose. In the pine stand, on the basis of studies conducted in the years 1995–1999 the proportion of stemflow in the inflow of water

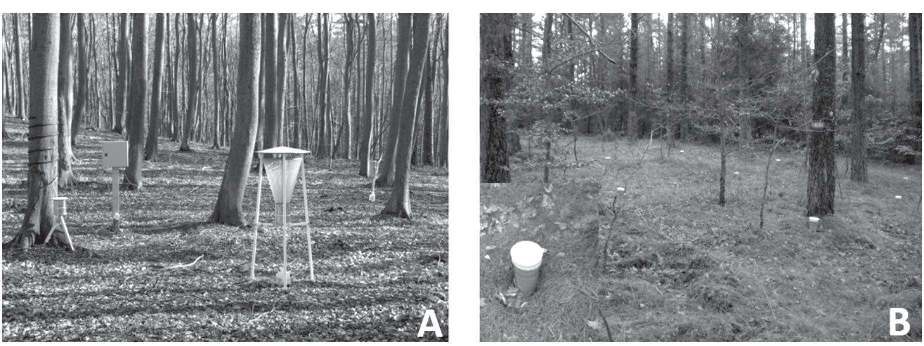

Photo 1. Throughfall collectors in the beech stand (A) and pine stand (B)

and dissolved substances to the forest floor was found to be negligible: the inflow of water averaged 0.2% (0.0–0.7) of the figure noted in the open (Kruszyk 2001). In the case of the loads of elements, the share of stemflow did not exceed 1% of total under-canopy deposition (TF+SF) (Kruszyk 2002).

Parallel to measuring deposition under the canopy, observations of the chemical composition of precipitation in the open close to the study areas were conducted. Precipitation measured in the open covered bulk precipitation (BP). Its collectors were exposed on a monthly or shorter basis if high precipitation totals occurred (in summer periods). In order to be able to compare the results, the collectors measuring throughfall, stemflow and precipitation in the open were exposed for identical periods. Both study areas belong to the measurement network of the Integrated Monitoring of the Natural Environment programme, a subsystem of the National Monitoring of the Natural Environment, with Storkowo (06ZM) and Wolin (11ZM) Base Stations.

The research involved an analysis of precipitation levels and basic physico--chemical parameters such as specific electrolytic conductivity (SEC), pH, and concentrations of dominant ions (chlorides, sulphates, nitrates, sodium, potassium, magnesium, ammonium and calcium). Precipitation amount, pH and specific electrolytic conductivity were measured for each collector separately on specified sampling dates. In the case of other parameters, their quantification was based on poured samples. Samples were poured proportionally taking into account the precipitation amount at individual positions. After filtration, they were stored and awaited analysis in a refrigerator at +4°C. The SEC values were automatically corrected to a standard value of 25°C. SEC and pH were measured using a multifunction meter (CX-741 made by Elmetron). Concentration levels of anions (sulphates, chlorides, nitrates) were determined using the ion chromatography method by means of a DX-120 chromatograph made by Dionex. Ammonium ions were determined colorimetrically using the Nessler method by means of a Spekol 1100 spectrophotometer made by Carl Zeiss. To measure concentration levels of sodium and potassium, atomic emission spectrometry was applied, and for those of calcium and magnesium – atomic absorption spectrometry. They were assayed using a SpectrAA-20 plus spectrometer made by Varian.

All chemical analyses were conducted at the Geoecological Station of the Adam Mickiewicz University located at Storkowo. Every year the Storkowo laboratory participates in inter-laboratory comparative studies of methods of assaying chemical compounds in water samples. These studies are intended to assess the correctness of assays in control samples derived from inter-laboratory comparisons organised under the EMEP network.

The *Statistica* v.12 program was applied for a statistical analysis of the obtained results, and *Grapher* v.9.0 for their graphic presentation. For every ion, its weighted mean was calculated, with precipitation or runoff levels taken as weights. Its load was obtained by multiplying its concentration by the amount of precipitation or runoff. As the available data series did not meet the assumption of the normality of distribution, non-parametric procedures were applied. To determine the statistical significance of differences between throughfall and bulk precipitation, the non--parametric Wald-Wolfowitz test was used.

In order to determine the range of precipitation transformation in the forest, an enrichment factor was applied (the rate of the load of an ion in precipitation under the canopy to its load in the open). All values exceeding 1 indicate an enrichment of throughfall in relation to precipitation in the open, all values below 1, a depletion of elements.

To determine the loads of ions (K^+, Ca^{2+}, Mg^{2+}) leached out of the canopy, the canopy budget model was used (Ulrich 1983; Bredemeier 1988; van der Mass, Pape 1991; Draaijers, Erisman 1995). It rests on two assumptions:

- sodium ions are not subject to any processes during infiltration of precipitation through the tree canopy, and
- particles containing ions: Ca^{2+}, Mg^{2+}, K^+ have the same size as particles containing Na^+.

These assumptions allow estimating the dry deposition factor DDF for those ions according to the formula (Ulrich 1983):

$$DDF = (TF_{Na} + SF_{Na} - BP_{Na})/BP_{Na}$$

where:
TF_{Na} – throughfall flux of sodium,
SF_{Na} – stemflow flux of sodium,
BP_{Na} – bulk precipitation flux of sodium.
Canopy leaching of these ions is calculated according to the formula:

$$CL_X = TF_X + SF_X - BP_X - DD_X$$

where:
DD_X – dry deposition flux.

For water, the following indicators were calculated: neutralisation of acidic compounds by calcium ions NF_{Ca} and ammonium ions NF_{NH_4} (Kulshrestha *et al.* 2003), as well as the proportion of acidic factors (Kostrzewski *et al.* 2007) using the following formulae (concentrations levels expressed in μeq·dm^{-3}):

$$NF_{Ca} = Ca^{2+} / NO_3^- + SO_4^{2-},$$
$$NF_{NH_4} = NH_4^+ / NO_3^- + SO_4^{2-},$$
$$\text{proportion of acidic factors} = NO_3^- / SO_4^{2-}.$$

Results

Throughfall expressed as a percentage of precipitation in the open is defined as tree stand permeability. This figure ranged from 60% in the examined beech stand to 70% in the pine stand. The proportion of beech stemflow in water reaching the forest floor was 7% of precipitation recorded in the open. Taking into account throughfall and stemflow, the amount of water penetrating into the soil in the beech stand accounted for 67% of direct precipitation; interception made up the rest (33%).

During the research period specific electrolytic conductivity in bulk precipitation ranged from 9.85 $\mu S\cdot cm^{-1}$ to 48.60 $\mu S\cdot cm^{-1}$ in the Gardno Lake catchment and from 6.96 $\mu S\cdot cm^{-1}$ to 52$\mu S\cdot cm^{-1}$ in the Czarne Lake catchment (monthly values) (Fig. 2). Over the entire period, its weighted average SEC values amounted to 16.5 $\mu S\cdot cm^{-1}$ and 16.3 $\mu S\cdot cm^{-1}$, respectively. Using the classification of precipitation contamination levels developed by Jansen *et al.* (Leśniok 1996), they can be classified as slightly elevated (15 $\mu S\cdot cm^{-1}$–30$\mu S\cdot cm^{-1}$). Throughfall is much more mineralised. SEC values in the pine stand exceeded twice the values recorded in bulk precipitation (37.9 $\mu S\cdot cm^{-1}$ as the average for 2012–2014) and more than four times those in the beech stand (70.4 $\mu S\cdot cm^{-1}$) (Fig. 2). The maximum SEC value (342.0 $\mu S\cdot cm^{-1}$) was recorded in the pine stand in November 2011; this was a month with extremely low total precipitation (4.7 mm). As for beech stemflow water, its SEC (with an average of 85 $\mu S\cdot cm^{-1}$) was close to throughfall mineralization.

Percentage shares of individual ions in the chemical composition of precipitation for 2012–2014 calculated on the basis of weighted mean annual concentration levels of ionic components expressed in $\mu eq\cdot dm^{-3}$ are provided in table 1. In both study areas, chlorides were predominant anions. As for cations, their chemical composition

Table 1. Percentage shares of ions in the chemical composition of bulk precipitation, throughfall and stemflow

Location	Precipitation	Chemical composition	
		anions	cations
Beech stand	bulk precipitation	**Cl^-** (49) > **SO_4^{2-}** (28) >**NO_3^-** (23)	**Na^+** (34) = **Ca^{2+}** (34) > Mg^{2+} (12) > NH_4^+ (10) > K^+(7) > H^+ (3)
	throughfall	**Cl^-** (44) > **NO_3^-** (36) > SO_4^{2-} (20)	**Na^+** (28) > **Ca^{2+}** (25) > **K^+** (22) > Mg^{2+} (13)> NH_4^+ (12) > H^+ (0)
	stemflow	**Cl^-** (46) > **SO_4^{2-}** (33) > **NO_3^-** (21)	**Mg^{2+}** (26) > **Na^+** (25) > K^+ (20) >Ca^{2+} (16) > NH_4^+ (12) > H^+ (1)
Pine stand	bulk precipitation	**Cl^-**(36) > **SO_4^{2-}** (32) = **NO_3^-** (32)	**NH_4^+** (30) > **Ca^{2+}** (29) > **Na^+** (22) > Mg^{2+}(8) > H^+ (6) > K^+ (5)
	throughfall	**Cl^-** (46) > **NO_3^-** (28) > **SO_4^{2-}**(26)	**NH_4^+** (35) > Ca^{2+} (20) > Na^+ (19) > K^+ (15) > Mg^{2+} (9) > H^+ (2)

Ions with shares exceeding 20% of $\mu eq\cdot dm^{-3}$ of their content in the group of anions and cations, respectively, are marked in bold

Source: authors' own study.

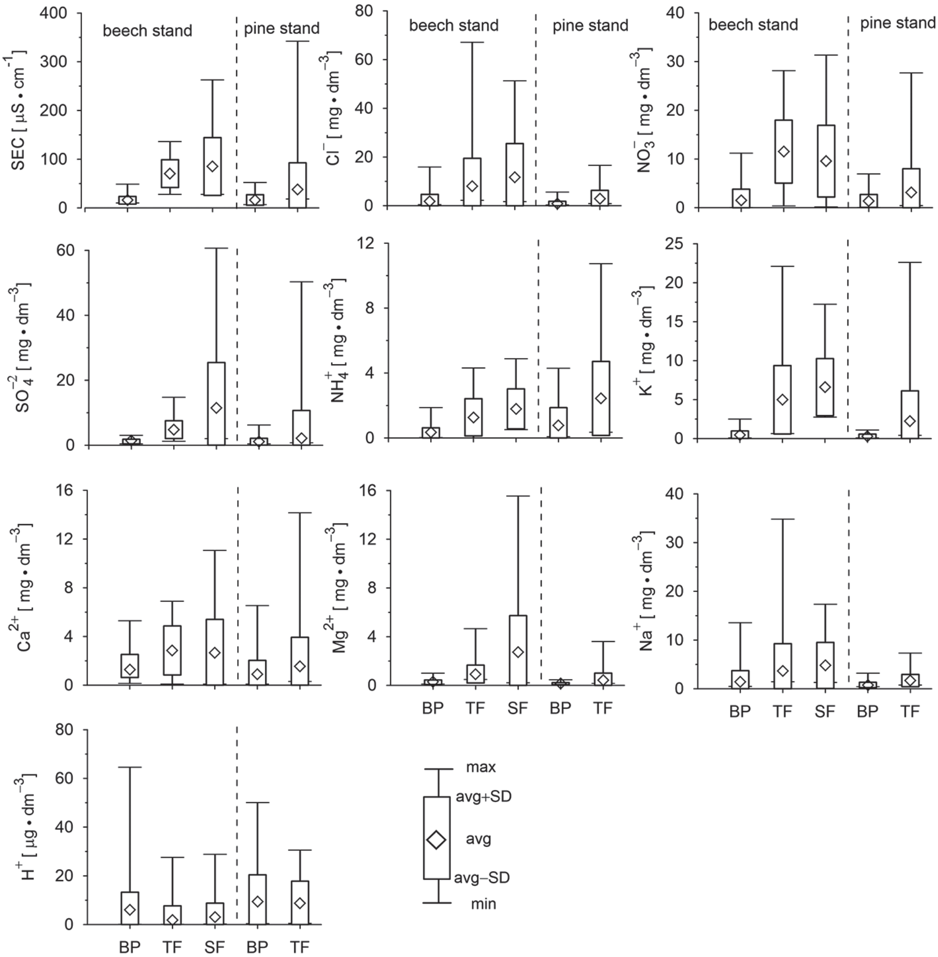

Fig. 2. Characteristics of the chemical composition of bulk precipitation (BP), throughfall (TF) and stemflow (SF) in the beech and pine stands

Source: authors' own study.

was different in the two areas. In the Czarne Lake catchment – both in BP and TF – the largest shares were those of ammonium ions and calcium. In the other catchment the proportions of cations were more diverse. Bulk precipitation was dominated by sodium and calcium, throughfall – by sodium, calcium and potassium, and stemflow – by magnesium and sodium.

For most ions in the Czarne Lake catchment, statistically significant dependencies of bulk precipitation and throughfall concentration levels were recorded. There were no such correlations for most ions in the other catchment (Tab. 2) apart from chlorides, sulphates, and potassium.

When comparing throughfall ion concentrations in the studied tree stands, the highest disparities were observed for chlorides, nitrates, sulphates and sodium. Their concentrations were higher in the beech stand. These differences were even wider for beech stemflow (Fig. 2). Only in the case of ammonium and hydrogen ions only were their annual average concentrations higher in the pine stand.

Table 2. Spearman's rank correlation matrix for bulk precipitation and throughfall concentration levels

Ion	Beech stand			Pine stand		
	N	r_s	α	N	r_s	α
Cl^-	32	**0.418**	0.017	31	**0.799**	0.000
NO_3^-	32	–0.172	0.346	31	**0.675**	0.000
SO_4^{2-}	32	**0.395**	0.025	31	0.284	0.121
Na^+	32	0.339	0.058	31	**0.671**	0.000
K^+	32	**0.585**	0.000	31	**0.580**	0.001
Mg^{2+}	32	0.314	0.080	31	**0.597**	0.000
H^+	32	0.218	0.232	30	**0.412**	0.024
Ca^{2+}	33	0.298	0.093	31	0.292	0.111
NH_4^+	32	0.255	0.159	30	**0.600**	0.000

N – number, r_s – Spearman's rank correlation coefficient, α – level of significance, correlation coefficients significant at $α \le 0.05$ are marked in bold

Source: authors' own study.

In spite of water loss via interception, the annual average load of ions under the canopy in both stands exceeded that in bulk precipitation (Tab. 3). This is the result of the ongoing process of leaching and wash-out of dry-deposited elements. Under the canopy of pines total deposition (68 $kg \cdot ha^{-1} \cdot year^{-1}$) was almost twice the amount of bulk deposition (36.7 $kg \cdot ha^{-1} \cdot year^{-1}$), while in the beech stand it was three times higher, at 52.6 $kg \cdot ha^{-1} \cdot year^{-1}$ and 160.2 $kg \cdot ha^{-1} \cdot year^{-1}$ (total throughfall and stemflow), respectively.

In both stands the highest enrichment as expressed by the enrichment factor was recorded for potassium, at 7.2 (beeches) and 5.6 (pines) (Fig. 3). Apart from potassium, high enrichment levels as compared with bulk precipitation were found for nitrates (5.0) and chlorides (3.0) in the beech stand and for ammonium ions (2.1), chlorides (2.3) and magnesium (2.1) in the pine stand. In both stands the enrichment of throughfall with chlorides was higher than with sodium.

Table 3. Annual deposition in the open and under the canopy [kg·ha⁻¹·year⁻¹]

	Period	Cl^-	NO_3^-	SO_4^{2-}	NH_4^+	K^+	Ca^{2+}	Na^+	Mg^{2+}	H^+*	Total
Beech stand	bulk precipitation										
	2012	14.21	12.23	12.17	2.99	1.97	8.63	13.93	2.45	82	**68.7**
	2013	6.85	8.15	7.11	2.02	3.69	5.24	4.57	1.04	10	**38.7**
	2014	12.86	7.29	6.92	0.93	3.09	9.70	7.91	1.61	12	**50.3**
	2012–2014	**11.31**	**9.23**	**8.73**	**1.98**	**2.92**	**7.86**	**8.80**	**1.70**	**35**	**52.6**
	throughfall + stemflow										
	2012	44.69	64.85	26.68	8.42	26.18	12.34	18.09	6.68	11	**207,9**
	2013	21.53	33.37	17.63	2.95	16.85	4.39	9.55	2.88	2	**109,2**
	2014	36.82	39.54	22.62	4.79	19.66	17.70	18.43	4.02	11	**163,6**
	2012–2014	**34.34**	**45.92**	**22.31**	**5.39**	20.90	11.48	15.36	4.53	8	**160,2**
Pine stand	bulk precipitation										
	2012	5.44	10.47	6.40	6.53	1.56	5.71	5.30	0.88	36	**42.4**
	2013	3.39	9.33	5.92	4.99	1.48	5.49	3.52	0.69	62	**34.9**
	2014	6.63	5.10	6.45	2.62	1.90	4.46	4.80	0.95	63	**33.0**
	2012–2014	**5.15**	**8.30**	**6.26**	**4.71**	**1.65**	**5.22**	**4.54**	**0.84**	**54**	**36.7**
	throughfall										
	2012	13.41	13.79	9.63	10.65	9.34	4.92	7.61	1.90	44	**71.3**
	2013	8.45	12.91	9.34	9.79	8.82	7.20	5.49	1.54	43	**63.6**
	2014	13.46	11.78	8.07	9.67	9.71	6.94	7.67	1.81	21	**69.1**
	2012–2014	**11.77**	**12.83**	**9.01**	**10.04**	**9.29**	**6.35**	6.92	1.75	36	**68.0**

* [g·ha⁻¹·year⁻¹]

Source: authors' own study.

This is also confirmed by Cl^-/Na^+ values. As for bulk precipitation in the Gardno Lake catchment, this ratio was 0.83, while in the Czarne Lake catchment it was equal to 0.74. As for throughfall, these values amounted to 1.43 and 1.1, respectively. In both stands the lowest enrichment figures (a factor below 2) were recorded for calcium and sodium. In the pine stand, also the sulphate and nitrate levels were close to those recorded in the open (Fig. 3).

On the basis of the three-year measurement series, a statistical testing procedure was employed to determine whether the apparent disparities between the deposition in the open and under the tree canopy were statistically significant. The non-parametric Wald-Wolfowitz test was applied for this purpose. For most ionic components the obtained differences in the deposition were statistically significant (Tab. 4). In both stands there were no significant differences in the deposition of sulphates, calcium, sodium and hydrogen ions as well as ammonium and magnesium ions under the canopy of beech trees.

The applied canopy budget model allowed calculating loads of potassium leached out of needles and leaves. In the pine stand the annual average deposition of potassium delivered with throughfall reached 9.29 kg·ha^{-1}·year^{-1}, and in the beech stand, also taking stemflow into account, it was equal to 20.9 kg·ha^{-1}·year^{-1}. The proportion of potassium leached out of plants in the total supply of this element to the soil amounted to 73.0% (6.78 kg·ha^{-1}·year^{-1}) in the pine stand and to 75.6% (15.80 kg·ha^{-1}·year^{1}) in the beech stand. Its inflow with bulk precipitation and through the wash--out of dry deposition reached 14.0% (2.92 kg·ha^{-1}·year^{-1}) and 10.4% (2.18 kg·ha^{-1}·year^{-1}), respectively. In the pine stand in the Czarne Lake catchment, these values were equal to 17.7% (1.65 kg·ha^{-1}·year^{-1}) and 9.3% (0.86 kg·ha^{-1}·year^{-1}).

The model also made it possible to calculate loads of calcium and magnesium ions leached out of needles and leaves. As for calcium, no leaching of this ion from plants was recorded. Its total load observed in the forest floor was linked with atmospheric deposition. This regularity occurs in both stands. Apart from potassium, leaching was also found for magnesium. In the beech stand loads of Mg^{2+} leached from leaves accounted for 34.4% (1.56 kg·ha^{-1}·year^{-1}) of the

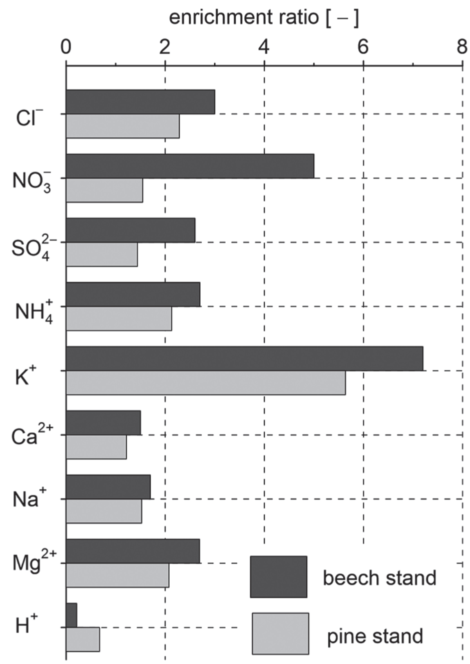

Fig. 3. Enrichment factor values for the beech and pine stands
Source: authors' own study.

Table 4. Results of the Wald-Wolfowitz test for the significance of differences in the loads in the open and under the canopy

Tree stand	H^+	Cl^-	NO_3^-	SO_4^{2-}	NH_4^+	Na^+	K^+	Mg^{2+}	Ca^{2+}
Pine	-	**	**	-	**	-	**	**	-
Beech	*	**	**	*	-	**	**	-	-

* difference significant at 0.05 < α ≤ 0.1; – difference significant at α > 0.1; ** difference significant at α ≤ 0.05

Source: authors' own study.

total deposition of this element under the canopy. In the pine stand this value was lower and amounted to 26.5% (0.46kg·ha^{-1}·year^{-1}). Bulk precipitation (48.2%) and the wash-out of dry deposition (25.3%) were the main routes of supply of this element to the ground in the pine stand. The proportions of these processes in the beech stand were 37.6% and 28.0%, respectively.

The weighted average annual pH of bulk precipitation for the period 2012–2014 was 5.25 in the Gardno Lake catchment and 5.02 in that of Czarne Lake. According to the classification of precipitation acidity developed by Jansen *et al.* (Leśniok 1996), in the Czarne Lake catchment bulk precipitation can be classified as having slightly reduced pH values (4.6–5.09), and in the Gardno Lake catchment – as standard precipitation. In the canopy, the pH of precipitation was higher: in the pine stand by 0.04 pH units (5.06), and in the beech stand by 0.48 pH units (5.73). Beech stemflow was more acidic than throughfall (pH = 5.51). The ratio of nitrates to sulphates (the indicator of the proportions of acidic compounds) in bulk precipitation amounted to 0.82 for the Gardno Lake catchment and 1.01 for that of Czarne Lake. In the neutralisation of acidic compounds in precipitation in the Gardno Lake catchment, calcium ions were more relevant than ammonium ions. Neutralisation indicators for NF_{Ca} and NF_{NH_4} amounted to 1.17 and 0.34 for BP, 0.5 and 0.25 for TF, and 0.34 and 0.25 for SF. In the Czarne Lake catchment, the proportions of those two ions in the neutralisation of acidic compounds in direct precipitation were similar, their NF_{Ca} and NF_{NH_4} indicators being equal to 0.99 and 0.96. As for throughfall in the pine stand, ammonium ions (NF_{NH_4} = 1.41) were more relevant in the process of neutralisation than calcium ions (NF_{Ca} = 0.8). In both stands the enrichment of their throughfall with H^+ ions was found to be below 1: in the beech stand it reached 0.2, and in the pine stand – 0.7. This demonstrates the neutralisation of acidifying substances in the canopy.

Discussion

Throughfall and stemflow (deciduous trees) are two of the supply routes of chemicals to the forest floor; their chemical composition is different from that of bulk precipitation. The increase in the mineralization of throughfall and stemflow results from the process of leaching of needles and leaves and washing dry deposition off tree surfaces (Likens, Bormann 1995; Grodzińska, Laskowski 1996; Hansen 1996; Małek , Astel 2008; Kozłowski *et al.* 2012). The capture of pollutants present in the air is particularly intensive in coniferous tree stands. It is caused by larger reception surfaces of needles and their presence throughout the year (Bochenek *et al.* 2008, Kozłowski 2003). The total load of ions reaching the forest floor with throughfall and stemflow in the beech tree stand was nearly 2.5 times higher than that observed under the canopy of pines (with a comparable level of deposition in the open). This is due, among others, to aerosols of marine origin. The study site in the beech stand is located on a cliff near the open sea. It also tends to have advective fogs which provide extra water supplies (Tylkowski, Samołyk 2012). The research conducted in the years 2009–2010 in the Gardno Lake catchment made it possible to estimate this extra fog-derived water at 15.4% of the total amount of water delivered to the ground in 2009 and 8.4% in 2010 (Tylkowski, Samołyk 2012). In individual months, fog water accounted for 0% to 52% of monthly water load delivered to the ground. Moreover, mist droplets contain pollutants at higher (up to tenfold) concentration levels than those observed in precipitation (Cape 1993; Kozłowski 2009). In the Gardno Lake catchment settlement misty show dozens of times greater mineralization compared to bulk precipitation, particularly high concentrations observed in the case of chlorides, sodium, sulphates and nitrates (Ścisłowska, Kostrzewski 2013).

Apart from ammonium and hydrogen ions, the average annual loads of other ions under the canopy of beech trees significantly exceeded those observed in the pine stand. The widest differences were recorded for nitrates (the load under the canopy of beech trees being more than 3.6 times higher), chlorides (2.9), magnesium (2.6), sulphates (2.5), potassium (2.2), and sodium (2.2), i.e. ions, apart from potassium, nitrates and sulphates, which originate chiefly from marine aerosols and atmospheric supply.

A relevant source of ions supplied to water in the canopy, apart from pollutants washed away from plant surfaces, is their leaching from leaves and needles. This concerns in particular such elements as potassium, magnesium, calcium and manganese. The model study of the beech and pine stands showed that in the case of potassium, canopy leaching accounted for 75.6% and 73%, respectively, of its total deposition on the forest floor. These values are close to those provided by Pajuste *et al.* (2006) for pine and spruce trees in Estonia (40%–73% of total deposition). Similar figures were also given by Herrmann *et al.* (2006) for two pine stands located in north-western Germany (44%–71%), but lower than those obtained by the present authors for oak trees (74%–92%). For uneven-aged spruce stands located in the Beskid Śląski Mountains, Małek and Astel (2008) received values ranging from 10% to 78%, depending on their age and the time of the year (growing period – non-growing period). For a beech stand located on the Święty Krzyż Mt. (Świętokrzyskie Mountains) Kozłowski *et al.* (2012) determined the proportion of potassium leaching at 91.9%, and for a fir stand, at 91.6%. The high proportion of potassium leached in the stands in the Świętokrzyskie Mountains can be linked with the high acidity of their throughfall. Its weighted average pH for a three-year study period was 4.43 for the beeches and 3.99 for the firs (Kozłowski *et al.* 2012). The intensity of potassium leaching depends on numerous factors, like the species and age of trees (Małek, Astel 2008; Kozłowski *et al.* 2012), canopy density (Whelan *et al.* 1998; Herrmann *et al.* 2006), distance from the forest margin (Devlaeminck *et al.* 2005), and inflows of acidifying compounds from the atmosphere (Draaijers *et al.* 1997; Polkowska *et al.* 2005; Małek, Astel 2008; Kozłowski *et al.* 2012). In the stands examined here, no calcium leaching from leaves or needles was recorded. The absence of this process can result from a deficiency of this element in the soil. The share of leaching in the supply of magnesium in both stands was lower than in the case of potassium, an element leached more easily because it is weakly bound to the structure of tissues (Wood, Bormann 1975). For both potassium and magnesium the percentage levels of their leaching in both stands were similar. The situation is different when the loads of leached ions are taken into consideration. In the beech stand the potassium load leached annually (15.47 $kg \cdot ha^{-1} \cdot year^{-1}$) was twice as high as that recorded in the pine stand (6.78 $kg \cdot ha^{-1} \cdot year^{-1}$). Even wider disparities occurred in the case of magnesium: over three times more of this element was leached in the beech stand. An analysis of leaching intensity over a hydrological year showed there to be seasonal variations. In both the pine and beech stands the maximum figures were recorded in the growing season, with a peak in September and October.

In both stands there was a decrease in the acidity of precipitation after it had passed through the tree canopy. In the pine stand H^+ loads accounted for 67.5% of bulk deposition, and in the deciduous stand for a mere 23%. This higher efficiency of the neutralisation of H^+ ions in deciduous stands should be associated with the leaching of alkaline ions being more intensive there than in coniferous stands.

Conclusions

The obtained results confirm that – in spite of reduced bulk precipitation under the tree canopy due to interception – the total load of elements supplied with throughfall to the ground during the study period exceeded the values recorded in the open. This regularity occurs in both, the coniferous and the deciduous tree stand.

Even though coniferous stands capture pollutants from the atmosphere more readily than deciduous ones, the observed deposition under the canopy was higher in the beech stand. On the one hand, this is due to the location of the beech stand near the Baltic Sea – a source of marine aerosols; and on the other hand, due to the presence of advective fogs which provide extra supplies of water and solutes.

The proportions of potassium and magnesium leaching in total deposition under the canopy in both stands were similar. There are differences when their loads are compared. In the beech stand the annually leached potassium load was more than twice as high as that recorded in the pine stand. The leaching process leads to a more intensive circulation of those elements in the ecosystem.

During penetration through the canopy precipitation water does not undergo acidification: the pH of throughfall in the examined stands was higher than of precipitation in the open. In both stands their H^+ enrichment factors were lower than 1. In the beech stand, it is calcium that plays a more important role in the neutralisation of acidic compounds, and in the pine stand, ammonium ions.

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