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Chemical composition of rainfall and throughfall under oaks, beeches and spruces in Kraków and Polanka Haller (southern Poland)

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Abstract – In 1990–1991 the total annual input of investigated ions (Ca^{2+}, Mg^{2+}, K^+, Na^+, NH_4^+, NO_3^-, Cl^-, SO_4^{2-}, PO_4^{3-}) with rainfall amounted to 16.0–18.7 g m^{-2} y^{-1} (a high percentage of sulphate ions in Kraków, and magnesium, potassium, phosphorus, and ammonium ions at Polanka Haller). The input of sulphur (1.98–3.35 g m^{-2} y^{-1}) was high in comparison with that noted in western and northern Europe. The input of nitrogen (0.44–0.58 g m^{-2} y^{-1}) was relatively small. The reaction of precipitations was not acid (pH value on average 6.3) owing to the neutralising effect of alkali metals. The sum of ions in throughfall under oaks and beeches was on average nearly double, and under spruces 3–5 times as great as in rainfall.

Key words: air pollution, sources of human activity, acid rain, chemical composition of rain.

Skład chemiczny opadów atmosferycznych na otwartej przestrzeni i pod koronami dębów, buków i świerków w Krakowie i Polance Haller (południowa Polska). W latach 1990–91 wraz z opadem atmosferycznym do gleby dostało się 16.0–18.7 g m^{-2} y^{-1} badanych jonów (Ca^{2+}, Mg^{2+}, K^+, Na^+, NH_4^+, NO_3^-, Cl^-, SO_4^{2-}, PO_4^{3-}). Dostawa siarki wyniosła 1.98–3.35 g m^{-2} y^{-1} — były to ilości wyższe niż notowane w Europie zachodniej i północnej. Dopływ azotu (0.44–0.58 g m^{-2} y^{-1}) był stosunkowo niski. Odczyn opadów atmosferycznych nie był kwaśny (średnio pH 6.3), co jest wynikiem jego neutralizacji przez pyły alkaliczne. Suma jonów w opadzie podkoronowym pod dębami i bukami była około dwukrotnie wyższa, a pod koronami świerków 3–5 razy wyższa, niż w opadzie na otwartej przestrzeni.

1. Introduction

One of the pathways of soil and surface water contamination by airborne pollutants is their dispersion by rainfall. The source of airborne mineral elements such as calcium, potassium, sodium and phosphorus are dusts originating from superficial layers of the soil and from industrial activity. Certain amounts of sodium and potassium are carried from the seas and oceans by winds. Gases such as SO_2, SO_3, NO_X, and dusts are by-products of industrial activity, which contaminate rainwater far away from the emitters. Contaminated rainfall may change the chemistry of ground water, especially the concentrations of chloride, sulphate, potassium, phosphorus, ammonium, and nitrate ions (Fiszer 1990). The results of investigations on the input-output budget in catchment areas in the Carpathian Mountains (E. Krzemien, A. Kurzbauer and J. Pawlik-Dobrowolski unpubl.) show that ammonium nitrogen and phosphorus as a rule formed a high percentage in the precipitation. The wash-out of dusts and gases from the air leads to a strong acidification of rainwater, though by enhanced loading of the air by dusts the fall in reaction value may be negligible owing to presence of alkaline compounds, including alkali metals (Na, K) and alkaline earth metals (Mg, Ca), efficiently neutralizing acids. Sulphates and nitrates of calcium, magnesium, sodium, and potassium are the products of this reaction.

The concentration of chemicals in atmospheric precipitation depends, inter alia, on the quantity of pollutants emitted by industrial centres. The purpose of these investigations was to compare the differences in concentration of chemical elements in rainfall (bulk precipitation) and in throughfall within and outside a large town.

2. Study area

The investigations were carried out in the Botanical Garden of the Jagiellonian University in Krakow (50°04' N, 19°58' E; alt. 206 m) and in the forest "Las Uniwersytecki" at Polanka Haller (49°56' N, 19°45' E; alt. 285 m), in an open area and under the canopies of three tree species: beech, *Fagus silvatica* L., oak, *Quercus robur* L., and spruce, *Picea abies* (L.) Karst. The Botanical Garden is located in the centre of Kraków hence air contamination is very high there. The nearest high emitters are: the steel works in the district of Nowa Huta, the thermal power station "Łęg", and the pharmaceutical factory "Polfa". The forest in the village Polanka Haller (c. 20 km SW of the centre of Kraków) is surrounded by arable land. A more detailed description of the site is given in Drużkowski et al. (1984). As westerly and south-westerly winds prevail in the investigated area, constituting altogether about 40% windy days, there is also an additional source of airborne pollutants owing to their long-distance transport from the industrial centres of Upper Silesia (southern Poland) and Ostrawa-Kamińsk (Czech Republic).

3. Material and methods

The chemical composition of precipitations was investigated in two spring–autumn periods (from April to October in 1990 and 1991) and one winter period (from November 1990 to March 1991). Water for chemical analysis was sampled every month using eight sample plots. Four of them were located in the Botanical Garden in Krakow (twelve points beneath the canopies and three in the open area), and four in the forest at Polanka Haller (twelve points beneath the canopies and three in the open area). In each sample plot three funnel and bottle assemblies (catchment area 125 cm^2 at the height of 35 cm) and one Hellman's collector were placed. In the open area were placed two bottles and one collector.

Each water sample was analyzed separately. The determinations of pH and concentrations of Ca^{2+}, Mg^{2+}, K^+, Na^+, NH_4^+, NO_3^-, Cl^-, SO_4^{2-}, and PO_4^{3-} ions were made. The following methods were used: calcium, potassium, and sodium were determined photometrically (Flapho-4), phosphates and nitrogen colorimetrically (Spekol), ammonium nitrogen using Nessler's reagent, nitrate nitrogen using of phenoldisulphonic acid, magnesium using the versenate method, chlorine by titration, and sulphates nephelometrically with barium chloride.

Data collected beneath the canopies (four samples) and in the open area (three samples) were averaged and analyzed using the analysis of variance. The total mean sum of ions was calculated by summing up all the ions investigated and their influx to the soil was estimated as a product of the concentration and the amount of rain.

4. Results

4.1. Rainfall and throughfall

The total annual amount of rainfall from April 1990 to March 1991 reached 610 mm in Krakow and 697 mm at Polanka Haller, about three-quarters of it occurring from April to October. Throughfall under the beeches reached in summer 68% and in winter 78% of bulk precipitations. This value for oaks was 67% in summer and 82% in winter, and for spruces about 50% for the whole year. In these estimations the input by stemflow was not attempted, assuming its negligible amount.

4.2. Chemical composition of rainfall

The concentrations of ions in the rainfall collected in Kraków and at Polanka Haller differed considerably (Table I). Concentrations of calcium ions and sulphates were twice as great in Krakow as at Polanka Haller. The concentrations of other elements were also as a rule higher in Kraków than at Polanka Haller, though statistically significant only for calcium and sulphate ions ($p < 0.01$). Between concentrations of these ions a strong correlation was found ($r = 0.84$, $p < 0.001$). Comparison of the chemical composition of the rainwater revealed a higher percentage of sulphate ions in samples collected in Kraków, and potassium, magnesium, phosphorus, and ammonium ions in those from Polanka Haller.

Seasonal fluctuations in ion concentrations were found. During the spring–autumn period higher concentrations of calcium, potassium, and sulphates were noted while during the winter ammonium, chloride, and sodium ions occurred in higher concentrations. The total amount of dissolved elements in open areas in the spring–autumn period was in Kraków about 50% higher than at Polanka Haller. When comparing ion concentrations in the rainwater in the two spring–autumn periods of 1990 and 1991, in most cases higher concentrations of all ions were found in 1990, in both Kraków and at Polanka Haller.

4.3. Concentration of ions in throughfall

The sum of ions beneath the canopies of beech and oak was nearly twice as great as that in rainfall, both in Kraków and at Polanka Haller (Table I). Under spruces this amount was more than three times as great in Kraków, and five times as great at Polanka Haller. Amongst anions, sulphate and chloride prevailed, and calcium and potassium amongst cations.

Beneath the canopies of beech and oak a twofold increase in concentrations of calcium, sodium, sulphur, and nitrogen, and a fivefold increase in potassium were noted in Kraków. At Polanka Haller the throughfall was also richer in ions, although the increase in calcium and sulphur was negligible, contrary to potassium, whose concentration increased markedly, similarly as in Kraków. The highest percentage of potassium was noted in rainwater under the oaks.

Table I. Concentration of ions (mean and range; mg dm^{-3}) in rainfall and throughfall in Kraków and in Polanka Haller from April 1990 to March 1991.

Ions	Rainfall		Throughfall					
			Beech		Oak		Spruce	
Kraków								
Ca^{2+}	8.1	1.8–14.0	17.0	6.2–47.0	16.1	3.7–39.8	26.6	6.4–85.2
Mg^{2+}	1.4	0.2–2.8	1.2	0.2–4.3	1.2	0.2–3.7	1.4	0.4–3.3
K^+	1.2	0–2.9	6.9	0.1–22.3	12.5	0.5–27.7	13.3	0.8–22.4
Na^+	1.3	0–3.0	1.6	0.3–6.6	2.0	0.3–4.3	2.6	0.4–9.0
NH_4^+	1.1	0.2–2.2	1.8	0.4–3.4	1.6	0.3–3.3	2.0	1.0–4.6
NO_3^-	0.7	0–2.2	2.6	0.7–2.8	1.2	0.4–2.7	1.7	0.4–3.7
Cl^-	16.1	6.3–28.4	29.2	13.8–62.0	26.2	13.2–46.7	52.1	14.2–106.2
SO_4^{2-}	17.6	1.8–38.3	26.6	5.2–50.0	36.8	4.5–98.0	59.1	4.1–188.2
PO_4^{3-}	0.05	0–0.20	0.76	0–0.55	0.04	0–0.30	0.03	0–0.04
Total	47.5	10.3–94.0	87.7	26.9–198.95	97.6	23.1–226.5	158.8	27.7–422.6
Polanka Haller								
Ca^{2+}	4.4	1.9–11.5	9.4	3.3–16.0	9.2	3.0–27.0	18.4	4.0–49.4
Mg^{2+}	1.1	0.2–2.8	0.8	0.1–3.0	0.9	0.05–1.6	1.4	0.1–3.8
K^+	1.5	0–2.7	6.2	0.3–49.0	9.3	0.3–38.6	17.7	0.1–59.8
Na^+	0.8	0–2.7	1.4	0.2–4.5	1.1	0.2–2.9	3.6	0.2–7.3
NH_4^+	0.9	0.4–2.0	1.8	0.6–5.3	2.1	0.7–5.8	3.7	0.5–9.5
NO_3^-	0.2	0–0.7	1.1	0.1–3.3	0.6	0.1–1.4	0.8	0.2–1.8
Cl^-	11.6	5.7–22.6	14.3	5.7–28.4	14.9	6.4–25.0	30.8	12.7–55.2
SO_4^{2-}	8.1	1.0–30.0	10.7	2.4–50.0	10.7	2.3–30.0	49.0	2.7–188.0
PO_4^{3-}	0.09	0–0.48	0.12	0.01–0.76	0.13	0.01–0.55	0.14	0.01–0.70
Total	28.7	9.2–72.8	45.8	12.7–160.3	48.9	13.1–132.8	125.5	20.5–375.5

The chemical composition of water collected beneath the canopies of spruces at Polanka Haller differed considerably from samples collected beneath beeches and oaks. The percentage of calcium was in this case twice that under beeches, and even six times that in the open area. Higher also were the concentrations of other elements. The amount of sulphur in rainwater collected beneath spruces in Kraków and at Polanka Haller was considerable higher than in the open areas, and higher than under beeches and oaks. The differences in concentrations of calcium, sulphate, and chloride ions in throughfall were statistically significant both between sites and among tree species.

4.4. Rainwater reaction

A high variability in the pH value of the rainwater reaction was noted during the study period. In the open areas it ranged from 5.1–6.9 and averaged 6.3 both in Kraków and at Polanka Haller. No significant correlation was found between the reaction of rainwater and the concentration of sulphate ions ($r = 0.42$, $p < 0.1$). Differences were also found in the pH value of rainfall and throughfall. Under spruces the pH of rainwater was the lowest among the tree species investigated and ranged from 4.7 to 6.5. The highest pH value was noted in rainwater collected under oaks (5.7–7.1). Rainwater under beeches had a slightly lower pH (5.2–7.0).

4.5. Influx of macroelements into the soil

From April 1990 to March 1991 the total (wet and dry) input of mineral elements (Ca, S, N, P, K, Mg, Na, Cl) into the soil reached 18.7 g m^{-2} y^{-1} in Krakow and 16.0 g m^{-2} y^{-1} at Polanka Haller (Table II). The bulk of the elements entered

Table II. The input of elements to the soil (g m^{-2} y^{-1}) with rainfall and throughfall in Krakow and Polanka Haller from April 1990 to March 1991.

Ions	Rainfall	Throughfall		
		Beech	Oak	Spruce
Kraków				
Ca	5.58	9.71	9.32	12.68
N+P+K	1.28	3.63	5.52	5.97
S	3.35	4.61	6.23	9.18
Mg	0.81	0.79	0.78	0.64
Na	0.59	1.08	1.31	1.40
Cl	7.14	9.35	8.31	13.40
Total	18.70	29.20	31.50	43.30
Polanka Haller				
Ca	3.29	2.44	2.67	10.12
N+P+K	1.93	2.04	4.71	8.40
S	1.98	1.57	17.80	71.60
Mg	0.70	0.61	0.46	0.74
Na	0.78	0.80	0.60	1.59
Cl	7.35	4.58	5.30	11.53
Total	16.00	12.00	15.50	39.50

the soil during the spring–autumn period, when the precipitation was higher. Among the total amounts of elements originating from the atmospheric precipitation calcium, sulphur, and chlorine had the highest percentage. The amount of elements deposited under the beech and oak in Kraków was on the average 1.5–2 times as great as in the open area but under the spruce measured values were even six times as great. There were no differences in the total input beneath broad-leaved trees and in the open area at Polanka Haller, only under spruce was the input about 2.5 times as great.

From April 1990 to March 1991 the influx of nitrogen to the soil from rainwater in Kraków reached 0.44 g m^{-2} y^{-1}, this amount being considerably higher under beeches (0.84 g m^{-2} y^{-1}), oaks (0.58 g m^{-2} y^{-1}), and spruces (0.64 g m^{-2} y^{-1}). At Polanka Haller the influx of nitrogen was greater than in Kraków and ranged from 0.58 g m^{-2} y^{-1} in the open area to 1.07 g m^{-2} y^{-1} under spruces. During the spring–autumn period an enrichment of the rainwater with nitrogen was observed under the canopies of trees in comparison with the open area; and on the other hand, there was a smaller amount of nitrogen under the trees than in the open area during the winter. During the study period the total influx of nitrogen, phosphorus, and potassium was estimated as about 5–10% of an average dose of the mineral fertilisers used in this area , i.e. 20.0 g m^{-2} y^{-1}.

5. Discussion

The concentration of sulphates in the precipitation during the study period (1.0–38.3 mg dm^{-3}) was higher than in western and in northern Europe, where the concentration of SO_4^{2-} ranges from 0.5 to 2.0 mg dm^{-3} (Siuta and Rejman--Czajkowska 1980). In other studies SO_4^{2-} precipitations of 12.8 mg dm^{-3} (E. Krzemień, A. Kurzbauer and J. Pawlik-Dobrowolski unpubl.), and of 18.5 mg dm^{-3} during the spring and 42.1 mg dm^{-3} in winter (Turzański 1991) were noted in Kraków. The concentration of calcium in rainwater found during the present studies was similar to that given by Denaeyer De Smet (1970) and Turzański (1991). An enhanced amount of mineral elements in precipitation is a common phenomenon in industrial regions. In Kraków and its environs Garścia and Sadowska-Janusz (1986) and Zając (1984) found considerable air pollution by dusts and gases. The results of phase analysis of dusts easily dissolved in water lead to the conclusion that almost all dust fall here is of industrial origin (Manecki et al. 1988). In Kraków the dusts from coal burning, metallurgic and calcium-dolomite prevail. Airborne dusts easily accumulate in rain drops and reach the soil surface. After three hours of rainfall only 35–55% of dusts remains in the atmosphere (Lisowski 1984). The high loading of rainwater by fertiliser elements (N, P, and K) in the forest at Polanka Haller results from agricultural activity in this area. The forest, in which the investigations were performed, is surrounded by arable land. The differences in concentrations of elements in rainwater in Kraków and at Polanka Haller result also from the various amounts of dusts easily dissolved in water, which is by 1/4 lower at Polanka Haller than in Kraków.

The great difference in the concentration of elements investigated in throughfall and in rainfall was probably the result of great dry deposition, especially in Kraków, which was then washed out by rain. The fall of dust in the vicinity of the Botanical Garden during the study period was estimated as 70.1 g m^{-2} (unpublished data from the Sanitary-Epidemiological Station in Kraków), and was considerably higher than the critical values for protected areas, i.e. 40 g m^{-2} (Ordinance 1990). In spite of this, trees growing in dense stands accumulated smaller amounts of dusts on leaf surfaces than scattered trees in the Botanical Garden in the centre of the town. The differences between coniferous and broad-leaved trees result from the fact that on the surface of needles dry deposition is accumulated throughout the whole year but on leaves only during the vegetation season.

The amount of nitrogen reached the soil with precipitation is an important component in the total input/output budget of mineral elements. According to Chojnacki and Kac-Kacas (1966) an annual influx of nitrogen into the soil with precipitation amounted to 1.0 g m^{-2} near Puławy (central Poland). Kwiecień (1986) estimated this amount as 7.6–2.08 g m^{-2} in the Rybnik industrial centre (southern Poland). Stachurski and Zimka (1984) found that the passing of nitrogen in water solution through canopies is not a static process. When the input of nitrogen from the atmosphere is low its absorption by the canopies is relatively small. Thus, the enrichment of rainwater with nitrogen originates mainly from phytophagous activity. As a result a positive balance is observed. On the other hand, when the input of airborne nitrogen is high its absorption gains an advantage over enrichment by phytophagous activity and in consequence the balance is negative. Kwiecień (1986) found that the amount of nitrogen in rainwater depends on dust fall. The author also found that even small barriers (vertical screens) cause an enhanced dust fall, and as a result, a more intensive influx of nitrogen. According to Velthorst and Breemen (1989), rainwater, after passing through canopies, may be enriched with nitrogen two to four times. This is an undesirable effect because plants acquire nitrogen also from rainwater, which in turn may increase the level of nitrates.

The amount of sulphur entering the soil with precipitation in western and north-western Europe is estimated as 0.4–1.6 g m^{-2} y^{-1} (Siuta and Rejman--Czajkowska 1980). In relatively unpolluted agricultural areas in Poland it amounted to 1.0 g m^{-2} y^{-1} (Chojnacki 1967a). The input of sulphur into the soil in the open area in Kraków during the study period was about three, and at Polanka Haller two times greater. In the years 1983–1986 (with high precipitation, especially in 1985) this amount was estimated as 4.46 g^{-2} y^{-1} (Drużkowski and Szczepanowicz 1988). The decrease in sulphur concentration in rainwater during the study period was probably brought about by a decrease in industrial production and, in consequence, a diminished emission of sulphur. The lower input of most elements in 1991 as compared with 1990 was caused by lower precipitation and lower ionic concentrations. The input of sulphur with precipitation in Kraków is similar to that found in Tarnów (3.38 g m^{-2} y^{-1}) but lower than in Katowice (5.66 g m^{-2} y^{-1}) and Opole (8.42 g m^{-2} y^{-1}) (Chojnacki 1970). Particularly large amounts of sulphur enter the soil beneath spruces. According to Block and Niesar (1989), in Germany throughfall input of sulphur under spruce canopies was 3.3 times as great as in the open area. The present author's findings are in accordance with these results.

It is believed that an annual loss of calcium by washing out in the soil amounts to 3.0–5.0 g m^{-2} (Chojnacki 1967b), which may be counterbalanced by input with precipitation. The input of calcium in Kraków during the study period was higher in comparison with that found in Germany (0.54–1.34 g m^{-2} y^{-1}; Wiedey 1987). The great amount of alkaline elements in rainwater in the Kraków area, including an especially high percentage of calcium, neutralizes the excess of sulphur in the atmosphere, then reaching the soil, hence the reaction of rainwater in Kraków was not acid during the study period. A higher pH value found under the broad-leaved trees was brought about by washing-out dusts from the leaf surfaces by rain. Drużkowski and Szczepanowicz (1989), while analysing the chemical composition of snow in Kraków, found a coincidence of calcium and sulphur loading. Taking into consideration the present emissions of sulphur, nitrogen, and heavy metal compounds this is of special importance. The lack of neutralizing substances in the dusts (as carriers of calcium) could lead to far-reaching negative effects in the environment therefore it is necessary to eliminate sulphur dioxide from industrial smoke.

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