Atmospheric Deposition and Canopy Interactions in Urban Scots Pine Forest

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Žaltauskaitė, J. and Juknys, R. 2007. Atmospheric Deposition and Canopy Interactions in Urban Scots Pine Forest. *Baltic Forestry*, 13 (1): 68–73.

The investigations of atmospheric deposition were carried out in 2002 in urban Scots pine (*Pinus sylvestris* L.) stand in second biggest Lithuanian city – Kaunas. The main aim of this study was to investigate atmospheric deposition and Scots pine canopy interactions at urban site and get more insight in dependency of precipitation chemistry on canopy closure. It was established that throughfall enrichment by anions was caused mainly by wash-off of dry deposited material, although there was a slight canopy exchange of chloride. By contrast, cations, and especially potassium enrichment came mainly from canopy leaching ($p<0.05$). Concentration of investigated ions in throughfall demonstrated a linear relationship with canopy closure. A twofold increase in canopy closure (40-80 %) led to an increase in the Cl^- ions concentration in throughfall by factor 3 and the concentrations of SO_4^{2-}, NO_3^- increased more than twice. The potassium concentration in throughfall grew by factor 2.4 along with the twofold increase in canopy closure.

Key words: bulk deposition, canopy closure, canopy interaction, Scots pine, net throughfall, throughfall

Introduction

Precipitation chemistry is modified as it passes through a forest canopy. Throughfall is enriched in some elements and depleted by others due to two primary processes: 1) wash-off of dry deposited compounds accumulated on the canopy between the rain events; 2) interaction between the forest canopy and rainfall – canopy exchange. Canopy exchange process includes leaching of elements (such as potassium, magnesium, calcium) from internal plant tissues and uptake of others (*e.g.* nitrogen) by foliage and epiphytic flora (Eaton *et al*. 1973, Lovett and Lindberg 1984, Lindberg *et al.* 1986, Schaefer and Reiners 1990).

As these throughfall deposition modification processes are closely related, it is very important to distinguish these processes because wash-off of dry deposition is attributed to an external input to the ecosystem, while leaching and uptake represent mostly an internal recirculation. Several attempts have been made to separate the contribution of these processes (wash-off, leaching, uptake) to the throughfall fluxes (Lovett and Lindberg 1984, Draaijers and Erisman 1995, Balestrini and Tagliaferri 2001).

The role of forest canopy in modifying the chemistry of precipitation has long been recognized. However, most studies on atmospheric deposition and canopy interaction have been focused on remote areas, whereas in urban areas few studies have been carried out (Sanusi *et al*. 1996, Lovett *et al*. 2000). Relatively few attempts have been made to account for canopy closure influence on throughfall patterns and little is known about quantitative relations between canopy closure and throughfall chemistry and deposition rates (Whelan *et al*. 1998, Bleeker *et al*. 2003).

Spatial variability of throughfall within stand is influenced by canopy structure. Field studies in coniferous stands have shown that throughfall fluxes decrease with the distance from the trunk (Pedersen 1992, Beier *et al*., 1993, Hansen 1996). Several studies have demonstrated close relationship between Leaf Area Index of the forest canopy and throughfall deposition (Stachurski and Zimka 2000, Zirlewagen and von Wilpert 2001). Observations at the Speulder Forest have shown that K^+ leaching increased with increasing canopy closure (Bleeker *et al*. 2003).

The main aim of this study is to investigate atmospheric deposition and Scots pine canopy interactions on urban site and get more insight in dependency of precipitation chemistry on canopy closure.

Materials and methods

The study was conducted on the urban Scots pine (*Pinus sylvestris L.*) stand situated approximately at 5km from the centre of the second biggest Lithuania city – Kaunas (55°42' N, 23°52' E). The study area was established in an even-aged, matured Scots pine stand, growing on podzolic soils. The stand is approximately 120 years old, 22-26 m high and the stand density is 260 trees ha^{-1}.

Collection of bulk and throughfall precipitation was performed on a monthly basis from April to November of 2002.

Twelve throughfall collectors were randomly installed in the area of 50 × 50 m. Bulk deposition was collected with three collectors in a clearing of the forest. The collectors for bulk deposition and throughfall were identical. The collectors consisted of a polyethylene funnel of 17 cm diameter connected to a 5 l polyethylene bottle. The storage containers were placed in the holes into the ground in order to avoid light-induced alterations of collected water and to keep lower temperature.

Samples were analysed at the Lithuanian Institute of Physics. Analysis of anions (SO_4^{2-}, NO_3^- and Cl^-) was performed by ion chromatography with conductivity detection. Sodium (Na^+), potassium (K^+) and calcium (Ca^{2+}) were determined by the atomic emission method, pH was measured potentiometrically. The quality of the analytical data was checked by the cation – anion balance. The data quality is assured according to the EMEP manual for sampling and chemical analysis (EMEP 1996).

Deposition fluxes were calculated as the product of volume-weighted concentration and precipitation volume for each collection period and sampler. Net throughfall fluxes were calculated by subtracting bulk deposition fluxes from throughfall fluxes. Significance of differences between bulk and throughfall deposition was assessed with Mann-Whitney test. Differences were considered statistically significant at $p<0.05$.

Canopy closure is an approximate indicator of stand density. It is expressed as a proportion or percent of the ground area covered by the vertical projection of the tree crowns above collector. The closure of canopy cover above each collector was estimated by image analysis of canopy photographs taken vertically from the centre of each collector (Fig. 1). Images were made with HP PhotoSmart C500 Digital camera, using a 28-mm standard lens pointing vertically, making sure the zenith angle of the camera equalled 0°. Images were compiled in a Geographic Information system (GIS) and analysed with the ArcView program. The number of black pixels divided by the total number of pixels was determined and used as an estimate of canopy closure above each throughfall funnel.

Figure 1. Examples of canopy cover above different throughfall funnel images

Results

Data on bulk and throughfall precipitation chemistry are presented in Table 1. Bulk precipitation was weakly acidic, pH ranged from 4.83 to 6.94. Most abundant anions in bulk precipitation (SO_4^{2-} and NO_3^-) were almost totally balanced (in equivalent basis) by NH_4^+ and Ca^{2+}. Mean amount of throughfall (31.23 mm $month^{-1}$) was statistically significantly less than the amount of bulk precipitation (42.88 mm $month^{-1}$) and it amounted only to 73 % of bulk precipitation.

The high filtering capacity of Scots pine canopy leads to the ionic enrichment of throughfall. Theconcentrations of all investigated ions in throughfall were found to be higher of that in bulk precipitation, though an increase in the concentrations of NH_4^+-N, Ca^{2+} and H^+ was statistically insignificant ($p>0.05$) (Table 1).

Table 1. Volume – weighted mean concentrations (μeq l^{-1}) and fluxes (mg m^{-2} $month^{-1}$) in bulk deposition and throughfall of the study period (April-November of 2002)

	SO_4-S	NO_3-N	Cl^-	NH_4-N	Na^+	K^+	Ca^{2+}	H^+
Concentrations								
Bulk	**48.7**	**53.6**	**20.6**	80.0	**21.7**	**25.5**	52.5	5.7
Throughfall	**84.4**	**101.4**	**42.2**	118.6	**29.1**	**67.5**	66.0	8.8
Fluxes								
Bulk	33.4	32.2	**31.3**	48	21.3	**42.7**	45	245.3*
Throughfall	42.2	44.4	**46.8**	51.8	20.9	**82.5**	41.2	274.8*
Net throughfall	8.8	12.2	15.5	3.8	-0.4	39.8	-3.8	29.5*

* - Units in μeq m^{-2}. Significant differences between bulk and throughfall are shown in bold characters (Mann-Whitney U-test)

Taking into account, that the concentrations of ions were higher in throughfall in comparison with bulk precipitation and that the amount of throughfall precipitation was essentially less than the amount of bulk precipitation, differences in bulk and throughfall fluxes were less expressed than these in the concentrations (Table 1). Statistically significant increase was detected only for K^+ and Cl^- fluxes. Ionic pool of sodium was almost unchanged after passing through the Scots pine canopy, and the fluxes of other analysed ions with throughfall were greater than with bulk precipitation, however this difference was statistically insignificant.

While for both inorganic nitrogen compounds (nitrates and ammonium) net throughfall fluxes were positive, *i.e.* throughfall was enriched by nitrogen (Table 1), though negative net throughfall fluxes, *i.e.* nitrogen canopy uptake was observed during some months (June, July) of the vegetation period. (Fig. 2). At the same time it is necessary to note that uptake of ammonium was greater than that of nitrates.

pine foliage prevails during the interaction of rainfall with tree canopy. K^+ throughfall fluxes were modified most of all and almost twofold increase in K^+ amount was detected comparing with fluxes in bulk deposition. Enrichment of throughfall in K^+ ions usually is explained by extremely high rate of potassium leaching from leaf tissue (Tukey 1970 cited in Moreno *et al.* 2001).

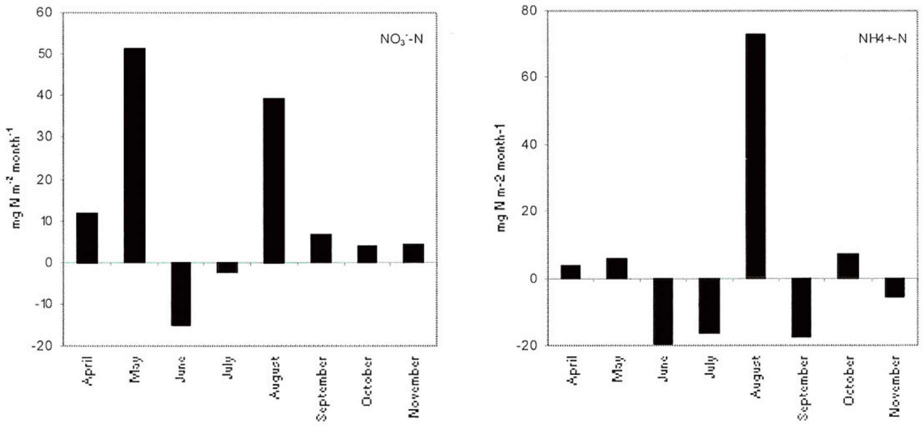

Figure 2. Monthly net throughfall fluxes of nitrates and ammonium (mg m^{-2}) (April-November of 2002)

To evaluate the relative contribution of all three rainfalls chemistry modification processes – wash-off, leaching and uptake, the following presumptions based on the findings of different investigators (Lovett and Lindberg 1984, Rodrigo *et al.* 2003) were used:

1. If net throughfall is independent of precipitation volume, the enrichment of throughfall is linked to dry deposition.

2. If net throughfall is positively related with precipitation volume, the ionic enrichment is resulted by leaching ions from plant tissue.

3. If net throughfall is negatively related with precipitation volume, the decrease in ions fluxes in throughfall is attributed to canopy uptake.

Ions of anthropogenic origin (SO_4^{2-}, NO_3^-) followed the first presumption, *i.e.* enrichment of throughfall by these ions could be attributed to dry deposition (Table 2). As studies were conducted in the urban forest stand it could be attributed to the proximity of local industry and traffic sources.

Table 2. Spearman correlation between net throughfall fluxes and amount of throughfall precipitation

	SO_4-S	NO_3-N	Cl^-	NH_4-N	Na^+	K^+	Ca^{2+}	H^+
r-value	0.09	0.17	0.24	-0.13	0.04	0.35	0.24	0.45
p-value	0.38	0.11	0.03	0.22	0.68	0.001	0.03	0.00001

Statistically significant positive correlation of K^+, Cl^- and H^+ net throughfall with the volume of precipitation indicate that leaching of these ions from Scots

Negative, though statistically insignificant correlation with throughfall volume was detected only for ammonium fluxes (Table 2) and it suggests uptake of ammonium by Scots pine canopy.

The effect of relative canopy closure on the variability of throughfall chemistry were investigated further. The concentrations of all ions in throughfall positively correlated with canopy closure. For sulphates, nitrates, chlorides and potassium the correlation was statistically significant (Table 3).

Table 3. Pearson correlation between canopy closure and concentration of investigated ions in throughfall (Statistically significant values are shown in bold characters)

	SO_4-S	NO_3-N	Cl^-	NH_4-N	Na^+	K^+	Ca^{2+}	H^+
r-value	**0.31**	**0.22**	**0.31**	0.04	0.2	**0.27**	0.06	0.2
p-value	**0.003**	**0.04**	**0.004**	0.7	0.057	**0.01**	0.6	0.08

As seen in Figure 3, the concentrations of investigated ions in throughfall demonstrated a linear relationship with canopy closure. Twofold increase of canopy closure (40-80 %) led to an increase in Cl^- ions concentration in throughfall by factor 3 and the concentrations of SO_4^{2-}, NO_3^- increased more than twice. The potassium concentration in throughfall grew by factor 2.4 along with twofold increase in canopy closure.

Our results suggest that the higher canopy cover because of higher filtering capacity tends to collect bigger amounts of dry deposition of anions (sul-

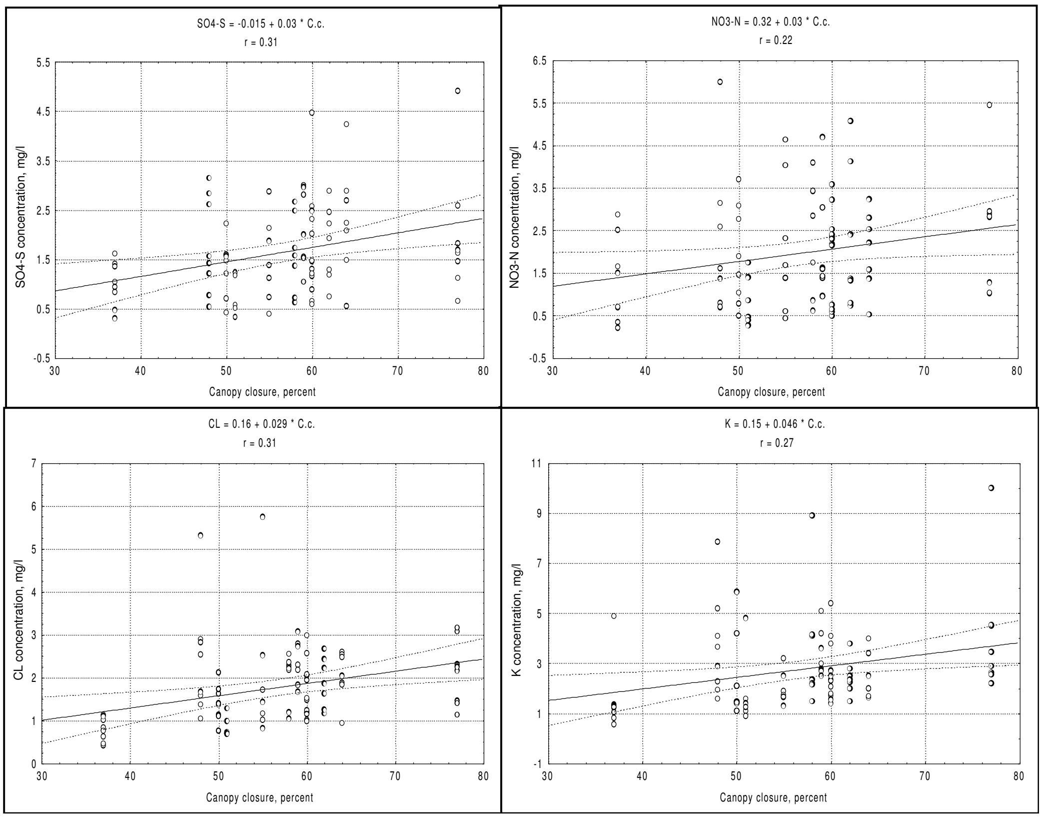

Figure 3. Relationship between ions concentrations in throughfall and canopu closure (C.c. - canopy closure)

phates and nitrates) as compared to smaller canopy cover. Bigger canopy cover also leads to higher rates canopy exchange and leaching of potassium.

Discussion

While analyzing atmospheric deposition and canopy interaction processes, generally it is accepted that sulphates in throughfall are derived only from atmospheric deposition, *i.e.* rainfall deposition, dry deposition of gaseous SO_2 and particulate SO_4^{2-}. Experiments with sulphur isotopes have shown that foliar leaching of sulphur is a minor contribution to enrichment of throughfall in SO_4^{2-} (Garten *et al.* 1988, Cape *et al.* 1992, Veltkamp and Wyers 1997), and it could be ignored. The results of our study showed statistically significant difference between the concentration of sulphates in throughfall and in bulk deposition (Table 1), however the correlation between net throughfall fluxes of sulfates and amount of throughfall precipitation was negligible (Table 2) and this led to conclusions that sulphur enrichment in throughfall was due to wash-off of dry deposited sulphur compounds.

Numerous studies have demonstrated that an essential part of atmospheric inorganic nitrogen deposition is absorbed by forest canopy and negative net throughfall usually is detected for both – oxidized and reduced nitrogen compounds (Lindberg *et al.* 1987; Lovett and Lindberg 1993, Wilson and Tiley 1998). Uptake of nitrogen has been widely reported in different forest types (Schaefer and Reiners 1990, Potter *et al*. 1991, Hansen 1996, Rodrigo *et al*. 2003). In our case, positive net throughfall of NO_3-N and NH_4-N for entire sampling period (April – November of 2002) was detected. It suggests that wash-off of dry deposition exceeds canopy uptake of nitrogen in urban forests, situated close to local pollution sources.

More detailed analysis of throughfall modification has shown that negative net throughfall fluxes, *i.e.* nitrogen canopy uptake, were observed during some months (June, July) of the vegetation period, indicating that canopy uptake exceeded external input of nitrogen compounds in these cases (Fig. 2). Increased nitrogen demand could lead to the increased uptake of nitrogen compounds during period of intensive growth. Other researchers (Hansen 1996) have obtained similar results that showed uptake in growing season and no uptake in dormant season.

According to our results in the period of more intensive canopy uptake ammonium was absorbed more efficiently than nitrate ions (57% vs. 37% in June and 48% vs. 6% in July). Different studies have shown that ammonium is taken up from both rain and gaseous – particulate fraction, and in case of nitrates uptake from rain is very negligible and occurs mainly from gaseous – particulate input (Lindberg *et al*. 1987; Stachurski and Zimka 2002). Another explanation of higher rates of ammonium uptake could be a negative charge of cuticle surfaces, therefore cuticle surface tend to repel anions such as nitrate, and attract cations such as ammonium (Schönherr 1982 cited by Wilson and Tiley 1998)

An enrichment of throughfall in K^+ ions usually is explained by high rate of potassium leaching from leaf tissue in association with acids (Tukey 1970 cited in Moreno *et al*. 2001) or through exchange with H^+ and NH_4^+ in leaf tissues (Shaefer and Reiners 1990, Moreno *et al*. 2001). As in this study there was no retention of H^+ (Table 1) and NH_4^+ retention was observed only in some periods of summer (Fig. 2), it suggests that K^+ ions were leached in association with acids.

Spatial variability of throughfall volume and fluxes within stand is determined by canopy structure. Field studies have usually demonstrated that tree species with dense canopies are more efficient filters of air pollutants (Ivens *et al*. 1990, Amezaga *et al*. 1997). Measurements made by Potter *et al.* (1991) showed that canopy cover accounted for 50-80 % of the variability in canopy exchange rates and for 60-70 % for the variability in dry deposition of cations. Meesenburg *et al*. (2005) found close relations between throughfall fluxes of nitrogen and stand volume, stand height and mean diameter, but no relation was found between nitrogen fluxes and canopy closure or LAI.

The results of our study have shown statistically significant dependence of most ions concentrations on canopy closure. The concentrations in throughfall demonstrated clear linear relationship with canopy closure (Table 3, Fig. 3). The results of our study were similar to those obtained by other studies there throughfall was analysed in relation to the distance from the stem. These studies have shown that ion concentrations and fluxes in throughfall decrease with the distance (Pedersen 1992, Beier *et al.* 1993, Hansen 1996). It suggests that this spatial variability of throughfall is determined by canopy architecture. Enhanced deposition near the stem may be explained by increased canopy surface area and higher amounts of dry deposition on the top of the tree (Beier *et al.* 1993). In the case of deeper foliated canopy the water flow is slower and this leads to longer contact between the water and foliage. Consequently, the canopy exchange increases and rainfall washes off bigger amounts of dry deposited material. Furthermore, bigger canopy cover because of higher filtering capacity tends to collect bigger amount of dry deposition of gaseous and particulate compounds as compared to smaller one. It coincides with the results of Spearman correlation (Table 2) showing that the increase in sulphur and nitrogen in throughfall was due to dry deposition.

The bigger canopy closure represents the bigger amount of cation exchange sites (Schaefer and Reiners 1990), it leads to higher rates of canopy exchange. It explains bigger canopy leaching of ions such as potassium (Table 3, Fig. 3). It corresponds to the results of Spearman correlation (Table 2) showing that the increase in potassium in throughfall was due to leaching from the foliage.

The study highlights the need for more profound studies at differently polluted sites in order to advance our knowledge in throughfall deposition modification processes and in dependency of precipitation chemistry on canopy closure.

Acknowledgement

The authors wish to thank the Lithuanian State Science and Study Foundation, which has funded the research described in this article.

Special thanks to Dalia Jasinevičiene for time-consuming laboratory analysis and her useful consultations.

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Received 13 March 2006
Accepted 17 May 2007

ВЗАИМОДЕЙСТВИЕ МЕЖДУ АТМОСФЕРНЫМИ ВЫПАДЕНИЯМИ И ПОЛОГОМ СОСНЫ ОБЫКНОВЕННОЙ В ГОРОДСКОЙ ОКРУЖАЮЩЕЙ СРЕДЕ

Ю. Жалтаускайте и Р. Юкнис

Резюме

Исследования химического состава атмосферных выпадений были проведены в спелом древостое сосны обыкновенной (*Pinus sylvestris* L.), в Каунасе в 2002. Основной целью данного иследования было изучение взаимодействия между атмосферными выпадениями и пологом сосны обыкновенной в городской окружающей среде и расширение наших знаний о зависимости химического состава осадков от плотности полога. Было установлено, что обогащение осадков анионами происходит из-за увеличения сухих выпадений, а обогащение осадков катионами (K^+)– вымыванием из внутренних тканей хвои (п < 0,05). Концентрация ионов в выпадениях показали прямую зависимость от плотности полога. Увеличивание плотности полога от 40 до 80 % обусловило двухкратное увеличение концентрации SO_4^{2-}, NO_3^- и K^+ и трехкратное увеличение концентрации Cl^-.

Ключевые слова: атмосферные выпадения, плотность полога, сосна обыкновенная, нетто выпадения, выпадения под пологом леса

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Collection of bulk and throughfall precipitation was performed on a monthly basis from April to November of 2002.

Twelve throughfall collectors were randomly installed in the area of 50 × 50 m. Bulk deposition was collected with three collectors in a clearing of the forest. The collectors for bulk deposition and throughfall were identical. The collectors consisted of a polyethylene funnel of 17 cm diameter connected to a 5 l polyethylene bottle. The storage containers were placed in the holes into the ground in order to avoid light-induced alterations of collected water and to keep lower temperature.

Samples were analysed at the Lithuanian Institute of Physics. Analysis of anions (SO_4^{2-}, NO_3^- and Cl^-) was performed by ion chromatography with conductivity detection. Sodium (Na^+), potassium (K^+) and calcium (Ca^{2+}) were determined by the atomic emission method, pH was measured potentiometrically. The quality of the analytical data was checked by the cation – anion balance. The data quality is assured according to the EMEP manual for sampling and chemical analysis (EMEP 1996).

Deposition fluxes were calculated as the product of volume-weighted concentration and precipitation volume for each collection period and sampler. Net throughfall fluxes were calculated by subtracting bulk deposition fluxes from throughfall fluxes. Significance of differences between bulk and throughfall deposition was assessed with Mann-Whitney test. Differences were considered statistically significant at $p<0.05$.

Canopy closure is an approximate indicator of stand density. It is expressed as a proportion or percent of the ground area covered by the vertical projection of the tree crowns above collector. The closure of canopy cover above each collector was estimated by image analysis of canopy photographs taken vertically from the centre of each collector (Fig. 1). Images were made with HP PhotoSmart C500 Digital camera, using a 28-mm standard lens pointing vertically, making sure the zenith angle of the camera equalled 0°. Images were compiled in a Geographic Information system (GIS) and

Figure 1. Examples of canopy cover above different throughfall funnel images

analysed with the ArcView program. The number of black pixels divided by the total number of pixels was determined and used as an estimate of canopy closure above each throughfall funnel.

Results

Data on bulk and throughfall precipitation chemistry are presented in Table 1. Bulk precipitation was weakly acidic, pH ranged from 4.83 to 6.94. Most abundant anions in bulk precipitation (SO_4^{2-} and NO_3^-) were almost totally balanced (in equivalent basis) by NH_4^+ and Ca^{2+}. Mean amount of throughfall (31.23 mm $month^{-1}$) was statistically significantly less than the amount of bulk precipitation (42.88 mm $month^{-1}$) and it amounted only to 73 % of bulk precipitation.

The high filtering capacity of Scots pine canopy leads to the ionic enrichment of throughfall. Theconcentrations of all investigated ions in throughfall were found to be higher of that in bulk precipitation, though an increase in the concentrations of NH_4^+-N, Ca^{2+} and H^+ was statistically insignificant ($p>0.05$) (Table 1).

Table 1. Volume – weighted mean concentrations (μeq l^{-1}) and fluxes (mg m^{-2} $month^{-1}$) in bulk deposition and throughfall of the study period (April-November of 2002)

	SO_4-S	NO_3-N	Cl^-	NH_4-N	Na^+	K^+	Ca^{2+}	H^+
Concentrations								
Bulk	**48.7**	**53.6**	**20.6**	80.0	**21.7**	**25.5**	52.5	5.7
Throughfall	**84.4**	**101.4**	**42.2**	118.6	**29.1**	**67.5**	66.0	8.8
Fluxes								
Bulk	33.4	32.2	**31.3**	48	21.3	**42.7**	45	245.3*
Throughfall	42.2	44.4	**46.8**	51.8	20.9	**82.5**	41.2	274.8*
Net throughfall	8.8	12.2	15.5	3.8	-0.4	39.8	-3.8	29.5*

* - Units in μeq m^{-2}. Significant differences between bulk and throughfall are shown in bold characters (Mann-Whitney U-test)

Taking into account, that the concentrations of ions were higher in throughfall in comparison with bulk precipitation and that the amount of throughfall precipitation was essentially less than the amount of bulk precipitation, differences in bulk and throughfall fluxes were less expressed than these in the concentrations (Table 1). Statistically significant increase was detected only for K^+ and Cl^- fluxes. Ionic pool of sodium was almost unchanged after passing through the Scots pine canopy, and the fluxes of other analysed ions with throughfall were greater than with bulk precipitation, however this difference was statistically insignificant.

While for both inorganic nitrogen compounds (nitrates and ammonium) net throughfall fluxes were positive, *i.e.* throughfall was enriched by nitrogen (Table 1), though negative net throughfall fluxes, *i.e.* nitrogen canopy uptake was observed during some months (June, July) of the vegetation period. (Fig. 2). At the same time it is necessary to note that uptake of ammonium was greater than that of nitrates.

pine foliage prevails during the interaction of rainfall with tree canopy. K^+ throughfall fluxes were modified most of all and almost twofold increase in K^+ amount was detected comparing with fluxes in bulk deposition. Enrichment of throughfall in K^+ ions usually is explained by extremely high rate of potassium leaching from leaf tissue (Tukey 1970 cited in Moreno *et al.* 2001).

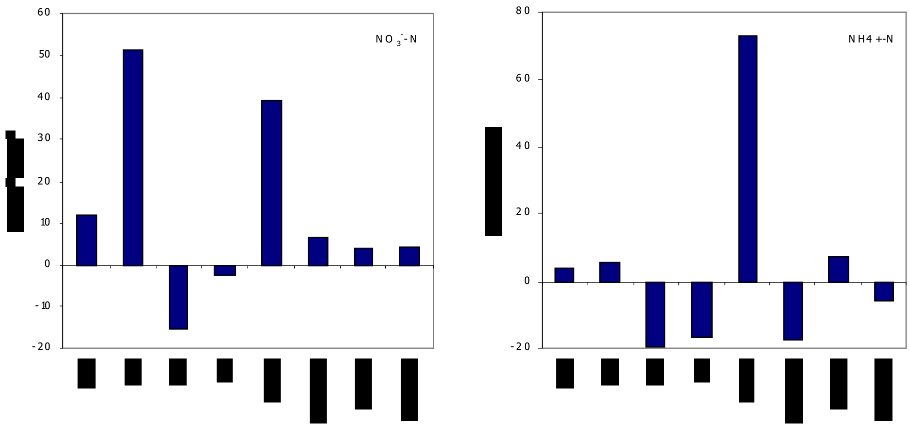

Figure 2. Monthly net throughfall fluxes of nitrates and ammonium (mg m^{-2}) (April-November of 2002)

To evaluate the relative contribution of all three rainfalls chemistry modification processes – wash-off, leaching and uptake, the following presumptions based on the findings of different investigators (Lovett and Lindberg 1984, Rodrigo *et al*. 2003) were used:

1. If net throughfall is independent of precipitation volume, the enrichment of throughfall is linked to dry deposition.

2. If net throughfall is positively related with precipitation volume, the ionic enrichment is resulted by leaching ions from plant tissue.

3. If net throughfall is negatively related with precipitation volume, the decrease in ions fluxes in throughfall is attributed to canopy uptake.

Ions of anthropogenic origin (SO_4^{2-}, NO_3^-) followed the first presumption, *i.e.* enrichment of throughfall by these ions could be attributed to dry deposition (Table 2). As studies were conducted in the urban forest stand it could be attributed to the proximity of local industry and traffic sources.

Table 2. Spearman correlation between net throughfall fluxes and amount of throughfall precipitation

	SO_4-S	NO_3-N	Cl^-	NH_4-N	Na^+	K^+	Ca^{2+}	H^+
r-value	0.09	0.17	0.24	-0.13	0.04	0.35	0.24	0.45
p-value	0.38	0.11	0.03	0.22	0.68	0.001	0.03	0.00001

Statistically significant positive correlation of K^+, Cl^- and H^+ net throughfall with the volume of precipitation indicate that leaching of these ions from Scots

Negative, though statistically insignificant correlation with throughfall volume was detected only for ammonium fluxes (Table 2) and it suggests uptake of ammonium by Scots pine canopy.

The effect of relative canopy closure on the variability of throughfall chemistry were investigated further. The concentrations of all ions in throughfall positively correlated with canopy closure. For sulphates, nitrates, chlorides and potassium the correlation was statistically significant (Table 3).

Table 3. Pearson correlation between canopy closure and concentration of investigated ions in throughfall (Statistically significant values are shown in bold characters)

	SO_4-S	NO_3-N	Cl^-	NH_4-N	Na^+	K^+	Ca^{2+}	H^+
r-value	**0.31**	**0.22**	**0.31**	0.04	0.2	**0.27**	0.06	0.2
p-value	**0.003**	**0.04**	**0.004**	0.7	0.057	**0.01**	0.6	0.08

As seen in Figure 3, the concentrations of investigated ions in throughfall demonstrated a linear relationship with canopy closure. Twofold increase of canopy closure (40-80 %) led to an increase in Cl^- ions concentration in throughfall by factor 3 and the concentrations of SO_4^{2-}, NO_3^- increased more than twice. The potassium concentration in throughfall grew by factor 2.4 along with twofold increase in canopy closure.

Our results suggest that the higher canopy cover because of higher filtering capacity tends to collect bigger amounts of dry deposition of anions (sul-

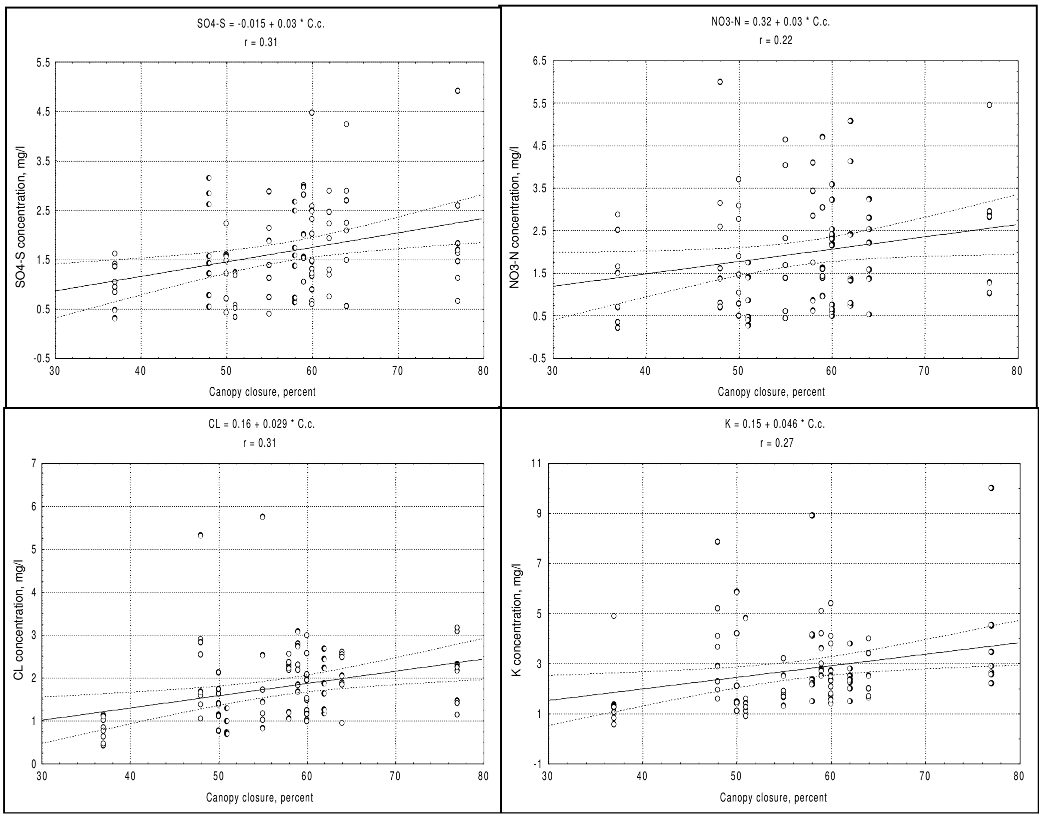

Figure 3. Relationship between ions concentrations in throughfall and canopu closure (C.c. - canopy closure)

phates and nitrates) as compared to smaller canopy cover. Bigger canopy cover also leads to higher rates canopy exchange and leaching of potassium.

Discussion

While analyzing atmospheric deposition and canopy interaction processes, generally it is accepted that sulphates in throughfall are derived only from atmospheric deposition, *i.e.* rainfall deposition, dry deposition of gaseous SO_2 and particulate SO_4^{2-}. Experiments with sulphur isotopes have shown that foliar leaching of sulphur is a minor contribution to enrichment of throughfall in SO_4^{2-} (Garten *et al.* 1988, Cape *et al.* 1992, Veltkamp and Wyers 1997), and it could be ignored. The results of our study showed statistically significant difference between the concentration of sulphates in throughfall and in bulk deposition (Table 1), however the correlation between net throughfall fluxes of sulfates and amount of throughfall precipitation was negligible (Table 2) and this led to conclusions that sulphur enrichment in throughfall was due to wash-off of dry deposited sulphur compounds.

Numerous studies have demonstrated that an essential part of atmospheric inorganic nitrogen deposition is absorbed by forest canopy and negative net throughfall usually is detected for both – oxidized and reduced nitrogen compounds (Lindberg *et al.* 1987; Lovett and Lindberg 1993, Wilson and Tiley 1998). Uptake of nitrogen has been widely reported in different forest types (Schaefer and Reiners 1990, Potter *et al*. 1991, Hansen 1996, Rodrigo *et al*. 2003). In our case, positive net throughfall of NO_3-N and NH_4-N for entire sampling period (April – November of 2002) was detected. It suggests that wash-off of dry deposition exceeds canopy uptake of nitrogen in urban forests, situated close to local pollution sources.

More detailed analysis of throughfall modification has shown that negative net throughfall fluxes, *i.e.* nitrogen canopy uptake, were observed during some months (June, July) of the vegetation period, indicating that canopy uptake exceeded external input of nitrogen compounds in these cases (Fig. 2). Increased nitrogen demand could lead to the increased uptake of nitrogen compounds during period of intensive growth. Other researchers (Hansen 1996) have obtained similar results that showed uptake in growing season and no uptake in dormant season.

According to our results in the period of more intensive canopy uptake ammonium was absorbed more efficiently than nitrate ions (57% vs. 37% in June and 48% vs. 6% in July). Different studies have shown that ammonium is taken up from both rain and gaseous – particulate fraction, and in case of nitrates uptake from rain is very negligible and occurs mainly from gaseous – particulate input (Lindberg *et al*. 1987; Stachurski and Zimka 2002). Another explanation of higher rates of ammonium uptake could be a negative charge of cuticle surfaces, therefore cuticle surface tend to repel anions such as nitrate, and attract cations such as ammonium (Schönherr 1982 cited by Wilson and Tiley 1998)

An enrichment of throughfall in K^+ ions usually is explained by high rate of potassium leaching from leaf tissue in association with acids (Tukey 1970 cited in Moreno *et al*. 2001) or through exchange with H^+ and NH_4^+ in leaf tissues (Shaefer and Reiners 1990, Moreno *et al*. 2001). As in this study there was no retention of H^+ (Table 1) and NH_4^+ retention was observed only in some periods of summer (Fig. 2), it suggests that K^+ ions were leached in association with acids.

Spatial variability of throughfall volume and fluxes within stand is determined by canopy structure. Field studies have usually demonstrated that tree species with dense canopies are more efficient filters of air pollutants (Ivens *et al*. 1990, Amezaga *et al*. 1997). Measurements made by Potter *et al.* (1991) showed that canopy cover accounted for 50-80 % of the variability in canopy exchange rates and for 60-70 % for the variability in dry deposition of cations. Meesenburg *et al*. (2005) found close relations between throughfall fluxes of nitrogen and stand volume, stand height and mean diameter, but no relation was found between nitrogen fluxes and canopy closure or LAI.

The results of our study have shown statistically significant dependence of most ions concentrations on canopy closure. The concentrations in throughfall demonstrated clear linear relationship with canopy closure (Table 3, Fig. 3). The results of our study were similar to those obtained by other studies there throughfall was analysed in relation to the distance from the stem. These studies have shown that ion concentrations and fluxes in throughfall decrease with the distance (Pedersen 1992, Beier *et al.* 1993, Hansen 1996). It suggests that this spatial variability of throughfall is determined by canopy architecture. Enhanced deposition near the stem may be explained by increased canopy surface area and higher amounts of dry deposition on the top of the tree (Beier *et al.* 1993). In the case of deeper foliated canopy the water flow is slower and this leads to longer contact between the water and foliage. Consequently, the canopy exchange increases and rainfall washes off bigger amounts of dry deposited material. Furthermore, bigger canopy cover because of higher filtering capacity tends to collect bigger amount of dry deposition of gaseous and particulate compounds as compared to smaller one. It coincides with the results of Spearman correlation (Table 2) showing that the increase in sulphur and nitrogen in throughfall was due to dry deposition.

The bigger canopy closure represents the bigger amount of cation exchange sites (Schaefer and Reiners 1990), it leads to higher rates of canopy exchange. It explains bigger canopy leaching of ions such as potassium (Table 3, Fig. 3). It corresponds to the results of Spearman correlation (Table 2) showing that the increase in potassium in throughfall was due to leaching from the foliage.

The study highlights the need for more profound studies at differently polluted sites in order to advance our knowledge in throughfall deposition modification processes and in dependency of precipitation chemistry on canopy closure.

Acknowledgement

The authors wish to thank the Lithuanian State Science and Study Foundation, which has funded the research described in this article.

Special thanks to Dalia Jasinevičiene for time-consuming laboratory analysis and her useful consultations.

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Received 13 March 2006
Accepted 17 May 2007

ВЗАИМОДЕЙСТВИЕ МЕЖДУ АТМОСФЕРНЫМИ ВЫПАДЕНИЯМИ И ПОЛОГОМ СОСНЫ ОБЫКНОВЕННОЙ В ГОРОДСКОЙ ОКРУЖАЮЩЕЙ СРЕДЕ

Ю. Жалтаускайте и Р. Юкнис

Резюме

Исследования химического состава атмосферных выпадений были проведены в спелом древостое сосны обыкновенной (*Pinus sylvestris* L.), в Каунасе в 2002. Основной целью данного иследования было изучение взаимодействия между атмосферными выпадениями и пологом сосны обыкновенной в городской окружающей среде и расширение наших знаний о зависимости химического состава осадков от плотности полога. Было установлено, что обогащение осадков анионами происходит из-за увеличения сухих выпадений, а обогащение осадков катионами (K^+)– вымыванием из внутренних тканей хвои (п < 0,05). Концентрация ионов в выпадениях показали прямую зависимость от плотности полога. Увеличивание плотности полога от 40 до 80 % обусловило двухкратное увеличение концентрации SO_4^{2-}, NO_3^- и K^+ и трехкратное увеличение концентрации Cl^-.

Ключевые слова: атмосферные выпадения, плотность полога, сосна обыкновенная, нетто выпадения, выпадения под пологом леса