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Throughfall Chemistry and Canopy Interactions in Urban and Suburban Coniferous Stands

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The research into atmospheric deposition by means of throughfall estimates were carried out in 2001-2004 in urban and suburban coniferous stands. Collection of bulk and throughfall deposition was performed on a monthly basis from April to November. The basic aim of the study was to investigate dry deposition contribution to total deposition at coniferous stands differently exposed to local pollution source. Bulk and throughfall deposition of anions in suburbs exceeded that in an urban site. Our results show that dry deposition is the main reason leading to enrichment of throughfall in ions of anthropogenic provenance (SO_4^{2-}, NO_3^-, Cl^- and NH_4^+). Regression analysis has revealed that the forest situated in the suburbs in the direction of the prevailing winds receive higher amounts of dry sulphur and nitrogen deposition than the forests growing in the city territories. It has been established that retained bigger amounts of anions deposition lead to the higher loss of base cations, especially of potassium.

Key words: *dry deposition, leaching, net throughfall, throughfall, uptake, urban-suburban.*

1. Introduction

Chemical composition of rainwater is strongly altered by the processes within the tree canopies before water reaches the forest floor as throughfall and may be enriched or depleted in different ions. Precipitation chemistry is modified because of two primary processes: 1) wash-off of dry deposited compounds (e.g., sulphur compounds) accumulated on the canopy between the rain events; 2) interaction between the forest canopy and the rainfall – canopy exchange. The canopy exchange process includes leaching of the elements (such as potassium, magnesium) from internal plant tissues and uptake of the others (e.g., nitrogen) by foliage and epiphytic flora (Lovett and Lindberg 1984; Lindberg et al. 1986; Schaefer and Reiners 1990). The tree canopy may act either as sink or source for various ions. Separation of the contribution of these three external and internal processes (wash-off, leaching and uptake) to the throughfall fluxes is very important and has been attempted by several researchers (Lovett and Lindberg 1984; Draaijers and Erisman 1995; Balestrini and Tagliaferri 2001).

There are two main ways to estimate dry deposition from measurements: micrometeorological methods and by means of throughfall. Dry deposition of gases and particles to forests is governed by the three main groups of factors: a) their concentrations in air and turbulent transport processes, b) chemical and physical nature or the deposition species, and c) capability of the underlying surface to capture or absorb them (Erisman, Draaijers 2003).

Cities and other local sources contribute highly to spatial variation of total atmospheric deposition and dry deposition as well. Emitting large amounts of acid precursors and dust to the atmosphere, cities can elevate concentrations of air pollutants and affect deposition rates, especially the rates of dry deposition, within hundreds of kilometres downwind of the city (Lovett et al. 2000; Chiwa et al. 2003).

The rate and pattern of atmospheric deposition in Lithuania are principally monitored under background pollution level, i.e., in Aukštaitijos and Žemaitijos National parks, in Preila (Augustaitis et al. 2005; Šopauskienė, Jasinevičienė 2006). Taking into account that most studies on atmospheric deposition have been focused on remote areas, whereas a few studies were concerned with urban and suburban areas (Juknys et al. 2007, Žaltauskaitė, Juknys 2007), the basic aim of this research has been to analyze the deposition fluxes and their modifications by forest canopy at two coniferous stands differently exposed to local pollution source (Kaunas city) and to evaluate relative contribution of dry deposition under different circumstances.

2. Materials and methods

Two mixed coniferous stands (Scots pine (*Pinus sylvestris* L.) and Norway spruce ((*Picea abies* (L.) H. Karst.)) growing at the sites differently exposed to the second biggest Lithuanian city – Kaunas have been chosen as the objects of this study. One sampling station, named urban, is in J.Basanavičiaus šilas in Kaunas city, approximately 5 km from the city centre (55°42′ N, 23°52′ E). The second sampling site, named suburban, is in the suburbs of Kaunas. The suburban site is located in Dubrava forest, where ICP Forest level II monitoring station 6M was established in 1990 (54°49′ N, 24°04′ E). The distance of this site is approximately 13 km from Kaunas centre and approximately 9 km from the urban site. 6M plot is established in mixed coniferous (pine-spruce) stand. Atmospheric deposition in that site was studied with researchers of the Forest Research Institute of Lithuania.

Collection of bulk and throughfall precipitation was performed on a monthly basis from April to November for four (2001-2004) years.

The studies (siting criteria, equipment, sampling periods, transportation and chemical analysis procedures) were conducted according to the ICP-Forests methods (UN/ECE, 1998). Bulk deposition was collected at each site with 3-4 collectors in a wind-shielded clearing of the forest. The areas of approximately 50 × 50 m were selected at each site and throughfall collectors were randomly installed (in J. Basanavičius šilas – n=23, in Dubrava forest – n=16). The storage containers of collectors were completely dark during the sampling period in order to avoid light-induced alterations of collected water and to keep the lower temperature.

Prior to analysis, the samples of bulk precipitation were combined to provide a composite one. Samples were analyzed 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 a cation – anion balance. The data quality was 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 the sites and between the bulk and throughfall deposition was assessed with Mann-Whitney U-test. Differences were considered statistically significant at $p<0.05$.

Regression and correlation analyses were used to determine the processes contributing to the deposition modification falling through the forest canopy.

3. Results and discussion

Data on mean monthly precipitation amount and volume – weighted mean concentrations of the principal ions in bulk and throughfall deposition are presented in Table 1.

Table 1. Mean monthly precipitation amount (mm) and volume – weighted mean concentrations (mg l^{-1}) of the ions in bulk and throughfall deposition (April – November of 2001 – 2004) at the study sites

	Precipitation amount	SO_4^{2-}-S	NO_3^--N	Cl^-	NH_4^+-N	Na+	K+	Ca^{2+}	H^+ *
Basanavičius stand									
Bulk	**46.15**	**0.69**	**0.76**	**0.61**	**0.91**	**0.47**	**0.76**	0.88	**3.71**
Throughfall	**35.46**	**1.22**	**1.25**	**1.95**	**1.32**	**0.78**	**2.71**	1.15	**5.64**
Dubrava forest									
Bulk	**57.06**	**0.73**	**0.84**	**0.77**	**0.66**	**0.55**	**0.62**	0.93	10.51
Throughfall	**38.30**	**1.34**	**1.50**	**2.16**	**1.12**	**0.66**	**4**	0.75	7.83

* - *µeq l^{-1}.*

Significant differences between bulk and throughfall are shown in bold characters (Mann-Whitney U-test).

During the studied period the mean monthly precipitation amount in the urban area was approximately by 19.2% lower than in the suburban area. Reduction in the precipitation amount in urban areas was reported by several authors (Sanusi et al. 1996; Lovett et al. 2000). More intensive air convection in urban territories and air pollution are considered as the main reasons of this phenomenon (Bukantis 1994; Rosenfeld 2000; Givati, Rosenfeld 2004).

The amount of throughfall was significantly lower than the amount of bulk precipitation due to the evaporation from leaf surface and canopy uptake. The difference was about 30%.

The concentrations of ions in bulk deposition in the suburban stand were higher than in the urban one. The mean concentration of Cl^- in suburban area was higher by 26%, NO_3^- - by 10.5%, SO_4^{2-} - by 6% that in the urban area, though the differences were statistically insignificant ($p>0.05$). Ammonium concentration was statistically significantly higher in J. Basanavičius stand in comparison to Dubrava forest.

As the pollutants' concentrations in precipitation decrease with the precipitation volume, the higher concentrations in the suburban area can not be explained by the impact of the precipitation volume, as the highest rainfall was detected there.

Thus, an increase in ion concentrations in the suburban site compared to the urban could be partially explained as a local effect from tall chimneys of power and industrial plants. Their emissions, sulphur and nitrogen oxides, first of all, result in local enrichment of precipitation by sulphates and nitrates mostly in suburban rather than in urban area.

High filtering capacity is characteristic of investigated Scots pine and Norway spruce stands leading to the ionic enrichment of throughfall. Concentrations of principal ions in throughfall were higher than those in bulk precipitation almost in all cases (Table 1). The differences in urban and suburban sites were similar: the concentrations of Cl^-, SO_4^{2-}, and NO_3^- in throughfall exceeded those in bulk precipitation by 2.8-3.2, 1.8 and 1.6-1.8 times, respectively. The highest enrichment was detected in the case of K^+ ions: in J. Basanavičius stand the concentration in throughfall the concentrations in bulk deposition exceeded by 3.6 times, in Dubrava stand – by 6.3 times.

The concentrations of ions in throughfall in suburban Dubrava stand were higher than in urban J. Basanavičius stand, though statistically significant difference was only in a case of potassium.

Data on deposition fluxes in the studied coniferous stands are presented in Table 2.

Table 2. Mean monthly bulk and throughfall deposition fluxes (April – November of 2001 – 2004) (mg/m²)

	Precipitation amount	SO_4^{2-}-S	NO_3^--N	Cl^-	NH_4^+-N	Na+	K+	Ca^{2+}
Basanavičius stand								
Bulk	**46.15**	32	34.9	**28**	42	21.9	**35**	41.6
Throughfall	**35.46**	39.6	40.7	**63.4**	42.9	25.2	**88**	37.9
Dubrava forest								
Bulk	**57.06**	41.6	47.9	**43.9**	37.7	31.2	**35.3**	**53.3**
Throughfall	**38.30**	51.3	57.5	**82.7**	42.8	25.8	**152.8**	**28.8**

Significant differences between bulk and throughfall are shown in bold characters (Mann-Whitney U-test).

Fluxes of all investigated anions were found to be significantly higher in the suburban area compared to the urban. Fluxes of most cations were also found to be the highest in the suburban area; however in most cases differences of the urban area were not statistically significant. Differences between suburban and urban fluxes with bulk precipitation were 30% for sulphates and 37% for nitrates. In the case of throughfall deposition, the highest fluxes were also observed at suburban Dubrava forest.

The enrichment of throughfall by most of the analyzed ions was observed. In the case of sulphur the enrichment was approximately 25%. For NO_3^- ions, the enrichment of throughfall in forests was about 20%, for ammonium – in suburban Dubrava stand enrichment was 13.5%, and in the urban forest there was no difference between bulk and throughfall deposition. Sodium, passing through the canopy, experienced minimal modifications and, on the contrary, potassium throughfall fluxes were modified mostly; 2.5- and 4.3-fold increase in K^+ amount was detected compared to the fluxes in bulk deposition in urban and suburban sites, respectively.

Net throughfall fluxes represent a final balance of all three precipitation modification processes – wash-off, leaching and uptake. Wash-off of dry deposition and leaching result in an increase in throughfall ion fluxes compared to the bulk deposition fluxes, whereas the uptake reduces throughfall fluxes and results in the negative net throughfall (Fig. 1).

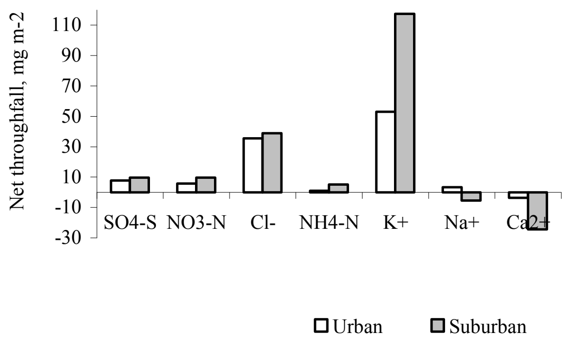

Fig. 1. Mean monthly net throughfall (mg/m²)

To evaluate a relative contribution of all three processes modifying precipitation – wash-off, leaching and uptake, the following presumptions based on the findings of different researchers (Lovett, Lindberg 1984; Balestrini, Tagliaferri 2001; Rodrigo et al. 2003) were used:

1. When net throughfall is positively related to the precipitation volume, the ionic enrichment results by ions leaching from a plant tissue.
2. When net throughfall is independent of the precipitation volume, the enrichment is linked to dry deposition.

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

The results of relationships between throughfall precipitation volume and net throughfall fluxes are shown in Table 3.

Table 3. Spearman correlation between the net throughfall fluxes and the amount of throughfall precipitation

	SO_4-S	NO_3-N	Cl^-	NH_4-N	Na^+	K^+	Ca^{2+}
J.Basanavičius s.							
r-value	0.07	-0.02	0.24	-0.15	-0.19	**0.42**	0.17
p- value	0.71	0.92	0.2	0.42	0.32	**0.02**	0.38
Dubrava forest							
r- value	-0.08	0.1	0.3	0.003	**-0.43**	**0.88**	**-0.46**
p- value	0.68	0.58	0.09	0.99	**0.01**	**0.000**	**0.01**

Significant values are shown in bold characters

Correlation between the net throughfall of SO_4^{2-}-S and the volume of precipitation was very weak and statistically insignificant and it proves that enrichment of throughfall by sulphates could be attributed to dry deposition. In the case of nitrogen (nitrates and ammonium), similarly to sulphates, there was found no more evident relations between the net throughfall and the amount of precipitation. These findings confirm that dry deposition is mostly responsible for throughfall modification.

At the urban site Cl^-, Na^+ and Ca^{2+} followed the second presumption, i.e. the enrichment of throughfall is linked to dry deposition. Canopy uptake could be attributed to Ca^{2+} and Na^+ at the suburban site. Uptake of calcium and sodium was observed in several studies (Draaijers et al. 1994; Amezaga et al. 1997).

Statistically significant positive correlation of K^+ net throughfall with the volume of throughfall was characteristic of the investigated sites and it confirms that leaching of this cation from Scots pine foliage prevails during interaction of rainfall with tree canopy. On the other hand, it could be seen that relationship was closer in a suburban site, it suggests that at the suburban site the leaching of potassium was more intensive than in the urban. Our results show that dry deposition is the main reason leading to enrichment of throughfall in ions of anthropogenic provenance (SO_4^{2-}, NO_3^-, Cl^- and NH_4^+). In order to find out whether the dry deposition contribution to throughfall is the same in forests in the city territory and in the suburbs in the direction of prevailing winds, we examined the relationship between bulk and throughfall deposition of sulphur and nitrogen compounds (Figs 2, 3).

The linear regression line between bulk and throughfall sulphate deposition shows a close relationship at both study sites (J. Basanavičius s. – R^2=0.55; Dubravos f. – R^2=0.49). From a quantitative point of view, however, we can observe some differences between the sites. For both sites (urban and suburban stands) the line of regression and many points were above y = x line, indicating throughfall enrichment. Moreover, at the suburban site coefficient b was higher than at the urban site (0.78 and 0.76), it suggests (Lovett, Lindberg 1984; Balestrini, Tagliaferri 2001) that at the suburban site dry deposition contributes more to total deposition.

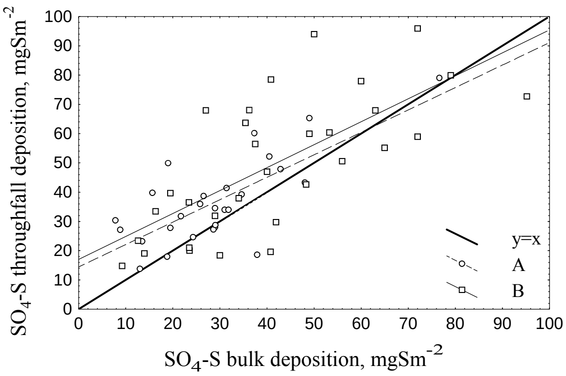

Fig. 2. Relationship between bulk and throughfall deposition of sulphur (A – J. Basanavičius s., B – Dubrava forest)

Similar results were obtained in a case of nitrates (J. Basanavičius s. – R^2=0.26; y=23.77+0.49*x; Dubrava f. – R^2=0.28; y=29.99+0.59*x) (Fig. 3). The results of regression analysis of ammonium fluxes are less statistically significant and more variable (J. Basanavičius s. – R^2=0.18; y=27.01+0.389*x; Dubrava f. – R^2=0.15; y=27.37+0.45*x) (Fig. 3), though coefficient b was higher in the case of suburb fluxes. It indicates that we can not distinguish one the main process by determining the load of ammonium ions as it is absorbed by the canopies and at the same moment may be washed-off from the foliage. As the interaction between ammonium and canopy is a very complicated process, we presume that intensity of the processes can not be explained by the analysis of a relationship between the throughafll fluxes and the precipitation amount or the fluxes in an open area.

Our results have led us to the conclusion that the forests situated in the suburbs in the direction of the prevailing winds receive higher amounts of dry sulphur and nitrogen deposition than the forests growing in the city territories.

The observations of G.M Lovett et al. (2000) in New York city and its surroundings at the distance of 11-128 km, revealed that the highest input of sulphur and nitrates was in the stand situated at approximately 15 km in the prevailing wind from the power station.

Positive correlation of K^+ net throughfall with the volume of throughfall indicates leaching of this cation from coniferous canopies. As the bulk deposition rates were very similar at the study sites and the relationship was closer in a suburban site, it suggests that at a suburban site the leaching of potassium was more intensive than in an urban site. In order to identify the reasons determining the higher leaching of potassium, we examined the relationship between the throughfall volume and the net throughfall fluxes of K^+ (Fig. 4).

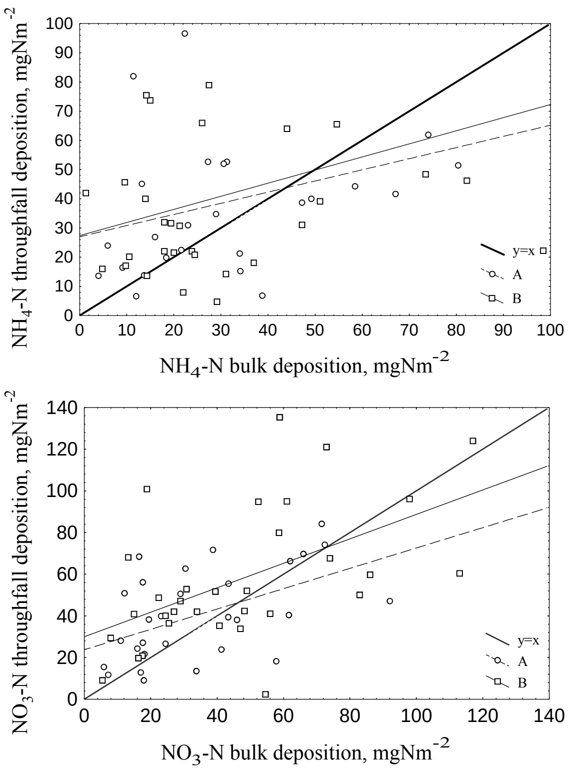

Fig. 3. Relationship between bulk and throughfall deposition of nitrogen compounds (A – J. Basanavičius s., B – Dubrava forest)

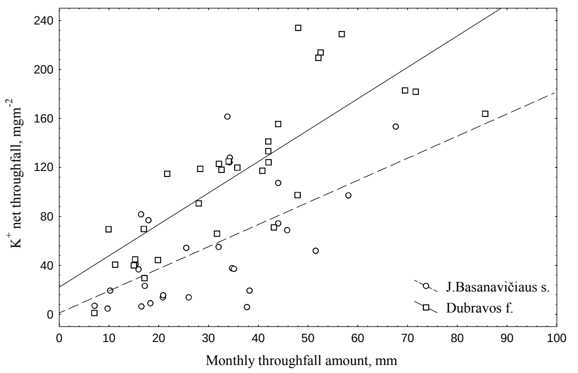

Fig. 4. Rrelationship between the amount of throughfall and the net throughfall of K^+ ions

The close relationship was determined at J. Basanavičius s. – R^2=0.35, b=1.8; Dubrava f. – R^2=0.63, b=2.56. Though comparing net throughfall of K^+ in the case of a mean amount of monthly precipitation (50 mm), leaching of potassium ions in a suburban area exceeds 1.7-fold leaching in an urban area, and these differences increase along with an increase in the amount of throughfall (Fig. 4).

Further investigations based on multiple regression analysis have shown that differences in a total amount of anions (SO_4^{2-} + NO_3^- + Cl^-) could be considered as the main reason resulting in differences in K^+ leaching at different sites (J. Basanavičius s. – R^2=0.35; Dubrava f. – R^2=0.75; $p < 0,05$). The throughfall amount and anion fluxes explained one third of the total K^+ leaching variation in J. Basanavičius s. and two thirds in suburban Dubrava forest. Thus, we can suggest that a higher dry deposition of ions of anthropogenic origin leads to higher leaching of cations. These results are in accordance with the findings of our previous study (Juknys et al., 2007).

4. Conclusions

Pollutants' fluxes were found to be greater in a suburban area situated in the direction of the prevailing winds. Differences between suburban and urban fluxes with bulk precipitation amounted to about one third for SO_4^{2-}, NO_3^-, Na^+, Ca^{2+}, and 1.6 times for Cl^-. Only in a case of potassium and ammonium the differences were negligible.

The atmospheric deposition changes essentially after the passage through the forest canopy. The statistical significant (p <0.05) enrichment of throughfall by most of the analyzed ions was observed, with an exception of sodium and calcium fluxes. The most essential enrichment took place in a suburban coniferous stand.

Dry deposition is the main reason leading to the enrichment of throughfall in ions of anthropogenic provenance (SO_4^{2-}, NO_3^-, Cl^- and NH_4^+). For nitrates ions, the enrichment of throughfall in urban and suburban pine stands was about 20%, for sulphates – about 25%. At a suburban site dry deposition contribution to the total deposition was bigger than in an urban site.

The study has shown that suburban forests situated in the direction of prevailing winds receive higher amounts of dry deposition of ions of anthropogenic provenance.

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Polajinių iškritų cheminė sudėtis ir jų sąveika su medžių lajomis spygliuočių medynuose, augančiuose mieste ir priemiestyje

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(gauta 2009 m. lapkričio mėn.; atiduota spaudai 2009 gruodžio mėn.)

Polajinių iškritų tyrimai buvo vykdyti 2001–2004 metais spygliuočių medynuose, augančiuose miesto ir priemiesčio sąlygomis. Atviros vietos ir polajinės iškritos tirtos balandžio–lapkričio mėnesiais. Pagrindinis tyrimo tikslas buvo ištirti sausų iškritų įtaką suminėms atmosferos iškritoms spygliuočių medynuose, augančiuose skirtingu atstumu nuo vietinės taršos šaltinio. Tyrimai parodė, kad sausos iškritos yra pagrindinė antropogeninės kilmės teršalų (SO_4^{2-}, NO_3^-, Cl^- ir NH_4^+) polajinių iškritų padidėjimą, palyginti su atviros vietos iškritomis, lemianti priežastis. Regresinė analizė parodė, kad vyraujančių vėjų kryptimi nutolusiame priemiestyje augančiuose medynuose sausų iškritų indėlis į sumines teršalų iškritas buvo didesnis nei mieste augančiuose spygliuočių medynuose. Nustatyta, kad didesnės anijonų iškritos lemia didesnį bazinių katijonų, ypač kalio, išplovimą iš medžių lajų.