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ORIGINAL PAPER

Pinus sylvestris and *Picea abies* canopy effects on deposition of air pollutants

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Abstract Tree canopies influence atmospheric pollutant depositions depending on type, ecosystem characteristics, and local climatic conditions. This study investigated the impact of *Pinus sylvestris* L. and *Picea abies* (L.) H. Karst., and a mixture of both, on the chemical composition of precipitation. Three permanent plots within the ICP forest level II monitoring network in Lithuania were selected to illustrate typical hemiboreal coniferous forests. The study analysed (1) the concentrations of NO_2, NH_3 and SO_2 in the ambient air; (2) the concentrations of SO_4^{2-}, NO_3^-, NH_4^+, Na^+, K^+, Ca^{2+} and Cl^- in throughfall beneath canopies and in precipitation collected in an adjacent field, and (3) S and total N, Na^+, K^+, Ca^{2+} and Cl^- depositions in throughfall and precipitation over 2006–2022. Results show a significant decrease in SO_2 emissions in the ambient air; NO_2 and NH_3 emissions also decreased. The canopies reduced the acidity of throughfall, although they led to notably higher concentrations of SO_4^{2-}, NO_3^-, Na^+, and particularly K^+. During the study, low variability in NO_3^- deposition and a decrease in NH_4^+ deposition occurred. Deposition loads increased by 20–30% when precipitation passed through the canopy. The cumulative deposition of S, Cl, Na, K, Ca, and N was greater under *P. abies* than under *P. sylvestris*. However, K deposition in throughfall was considerably lower under *P. sylvestris* compared to the *P. abies* or mixed stand. Throughfall S depositions declined across all three coniferous plots. Overall, there was no specific effect of tree species on throughfall chemistry.

Keywords Precipitation · Throughfall · Deposition of chemicals · Pollution · Lithuania

Introduction

The increase in human activities has led to higher emissions of air pollutants in urban areas, with some of these pollutants spreading to distant regions as background pollution (Kopáček et al. 2016). The effects of nitrogen (N) and sulphur (S) deposition on forest ecosystem services and the forest's role in enhancing air quality have been research topics for several decades (Galloway et al. 2008; Aherne and Posch 2013; Posch et al. 2019; Grennfelt et al. 2020; Costa et al. 2022).

Tree canopies have several significant effects on atmospheric depositions. These effects vary depending on pollutant type, ecosystem characteristics (species composition, age, stand density, canopy structure), and local climatic conditions (wind speed and direction, temperature, precipitation, movement of air masses) (Bredemeier 1988; Ponette-González et al. 2010; Tan et al. 2018). Airborne particles like pollen and fine particulate matter (PM) are captured by canopies as leaves and branches function as natural filters (Hamdan and Schmidt 2012; Corti et al. 2019). Trees also absorb specific gaseous pollutants, among which carbon dioxide (CO_2) should be mentioned first because it is directly involved in photosynthesis. Although trees use CO_2 for growth, they also absorb gases such as sulphur dioxide (SO_2), ozone (O_3), and nitrogen dioxide (NO_2), thereby reducing their concentration in the atmosphere. In open

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areas, urban settings, and forest environments, distinctions in atmospheric deposition become apparent due to the wind protection afforded by forests or groups of trees. This protection significantly diminishes the dispersion of various substances. Therefore, several studies suggest that forest canopy interception of precipitation is important for reducing atmospheric deposition of pollutants, trace gas fluxes and leaching of solutes (Whelan and Anderson 1996; Kozłowski et al. 2020).

Forest canopies absorb dissolved substances such as NH_4^+–N and NO_3^-–N in throughfall (Zhang and Liang 2012; Su et al. 2019). Although some studies have shown that canopies effectively filter trace metals, sulphur, and chlorine from precipitation, throughfall inputs can also change nutrient concentrations in the forest floor and soil (Zhao et al. 2017; Tan et al. 2018). Chemical transformations in air pollutant composition take place within the tree canopy, leading to substantial modifications in the chemical composition of precipitation before it reaches the forest floor, resulting in enrichment or loss of various ions (Germer et al. 2007; Hamdan and Schmidt 2012; Eisalou et al. 2013; Sheng et al. 2022). Two primary processes, the wash-off of dry deposited compounds which accumulate in the canopy between rain events, and ion exchange within the canopy are often highlighted (Žaltauskaitė and Juknys 2009; Tan et al. 2018; Sheng et al. 2022). High to moderate nitrate deposition in throughfall was found in central Europe, including Germany, Czechia, Poland, Austria, Italy, Slovenia and Belgium (Michel et al. 2022). However, high to moderate ammonium throughfall deposition was recorded in the larger area of central Europe than for nitrate. Calcium (Ca) and magnesium (Mg) depositions buffer acidifying effect in soil and high values were reported mainly in central and southern Europe (Michel et al. 2022). According to Schwartz et al. (2022), stands composed of conifers, hardwoods, and mixed species did not influence throughfall depositions.

The United Nations Economic Commission for Europe (UNECE), the Convention on Long-range Transboundary Air Pollution (Air Convention), and the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests) emphasised the importance of in situ measurements because of the notable local variability in pollutant deposition (Michel et al. 2022). This study aimed to determine if the canopies of coniferous species—*Pinus sylvestris* L., *Picea abies* (L.) H. Karst. and a mixture of both—change the chemical composition of precipitation. It was hypothesized that depositions in throughfall may be species dependent. In addition, the objective was to evaluate recent trends in elemental depositions, focusing on both precipitation which refers to rain/snow falling directly to the ground, and throughfall which is a portion of precipitation reaching the ground after interacting with and passing through the tree canopy.

Materials and methods

Study area

The study was carried out in Lithuania where forests cover over 33.8% of the total land area (State Forest Service 2022). Lithuania includes moderate lowlands and highlands with the highest mean elevation of 297.8 m a.s.l. The climate is moderately warm, passing from maritime to continental (Galvonaitė et al. 2013). According to the 1991–2020 climate normal, in Lithuania, the average annual temperature is 7.4 °C, and the average annual precipitation is 695 mm. The hottest month is July, and the coldest January. The highest precipitation of about 800 mm, is recorded in western Lithuania and determined by the proximity of the Baltic Sea and the specific terrain conditions. The highest annual global solar radiation of 3690 MJ m^{-2} is observed in South-Western Lithuania, while the lowest solar radiation of 3520 MJ m^{-2} is in south-eastern Lithuania. The dominant forest soils are sandy Arenosols and fertile Luvisols, with Cambisols predominate in agricultural lands (IUSS Working Group WRB 2015; Armolaitis et al. 2022).

Three study plots belonging to the ICP Forest level II monitoring network were selected (Fig. 1, Table 1).

The plots represent typical hemiboreal forests of coniferous stands, covering 56% of the total forest area. The three plots were set up within homogeneous stands, situated at a distance from urban centres, and considered representative of the regional environmental climate. The first study site, coded as a 3 M plot, was in the Kazlų Rūda forest approximately 29 km from Kaunas City (Table 1). This 3 M plot represents a Scots pine (*P. sylvestris*) stand. The second study site—a 6 M plot—was in the Kaunas district, 17 km from Kaunas centre. It represents a mixed Scots pine and

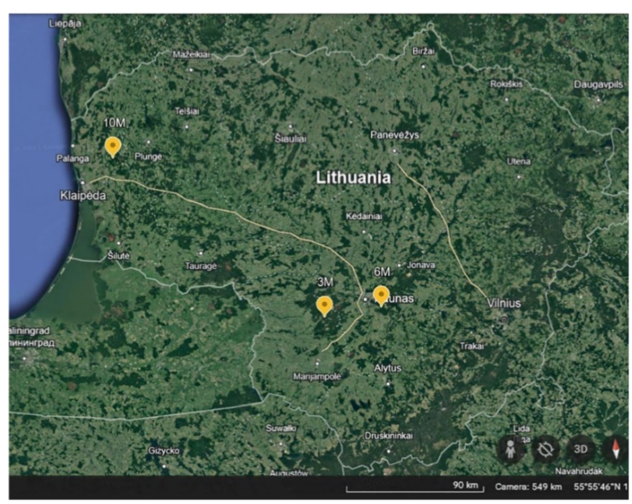


Fig. 1 Location of the ICP-forest level II monitoring plots (Lithuania)

Table 1 Characteristics of the permanent plots

Study plot	Plot code[a]	Latitude	Longitude	Established	Distance from city	Plot area (ha)	Forest site[b]	Soil type[c]	Vegetation type	Tree species composition	Age (years)[d]	Mean (cm)[e]	*DBH* mean growing stock volume (m^3 ha^{-1})[e]
Pinus sylvestris	3 M	54°46′608	23°34′864	Jun-95	29 km, Kaunas	0.25	Nb	Haplic arenosol	*Vacciniosa*	99% *P. sylvestris,* 1% *Betula* sp.	66	23.2 ± 0.5	444.7
Picea abies	10 M	55°49′780	21°28′199	Nov-06	24 km, Klaipėda	0.24	Lc	Gleyic luvisol	*Myrtillo-Oxalidosa*	100% *P. abies*	53	27.1 ± 0.5	298.7
Mixed forest	6 M	54°48′936	24°04′983	Jun-90	17 km, Kaunas	0.25	Lc	Haplic podzol	*Myrtillo-Oxalidosa*	74% *P. sylvestris*, 23% *P. abies,* 3% *Betula sp.*	100	23.2 ± 0.7	550

[a]According to the Lithuanian ICP-forests level II plots network

[b]According to the lithuanian forest site type classification, Nb is normal moisture oligotrophic soil, and Lc is temporarily over-moisture mesoeutrophic soil (Vaičys et al. 2006)

[c] Soil classified according to the world reference base for soil resources 2014 (IUSS Working Group WRB 2015)

[d]For plots 3 M and 10 M, tree age is for 2022; for plot 6 M, tree age is for 2017 because this plot was destroyed by wind and closed due to damage, the data sequence ends in 2017

[e]For plots 6 M and 10 M, the DBH and growing stock volume are given for 2015 and the 3 M for 2020

Norway spruce (*P. sylvestris* and *Picea abies*) stand. Data for this plot is accessible up to 2017, as observations were discontinued after wind damage in the same year. The third study plot—a 10 M plot—was in the Kretinga forest and represents a pure Norway spruce (*Picea abies*) forest. This study site was approximately 24 km from Klaipėda City. The forest site type is normally a moisture nutrient poor soil (Nb) in the first study site and temporarily waterlogged fertile soil (Lc) in the second and the third sites according to Lithuanian soil classification (Vaičys et al. 2006).

Sampling and analysis

Concentrations of nitrogen dioxide (NO_2), ammonia (NH_3) and sulphur dioxide (SO_2) in the atmosphere were determined with passive samplers on all plots from May to October. Two replicates of the passive samplers were exposed 2-m above ground level for 30 d in the open field (Schaub et al. 2016). The tubes were protected from sunlight by opaque cylindrical boxes. The samplers were analysed by PASSAM AG (Switzerland). NO_2 and NH_3 were determined using the photometric method, and SO_2 by ion chromatography.

Atmospheric deposition was sampled following the methodology of the ICP Forests monitoring program (Clarke et al. 2016). Sampling locations were beneath the tree canopy to capture throughfall and in an adjacent open field to collect precipitation across the three permanent plots.

Throughfall and precipitation samples were collected monthly during 2006–2020. Each plot was equipped with 16–18-cm diameter precipitation collectors positioned one meter above the ground. To avoid the impact of direct sunlight and temperature, the collectors were placed in black polyethylene pipes. Collectors damaged by human, animal, or other factors were renewed to ensure accuracy. Precipitation was sampled from three collectors installed in the open field 500 m from the forest plot. Throughfall samples record wet deposition, which involves determining the quantity of pollutants transported by rain and snow. Additionally, throughfall samples include dry deposition from particulate matter accumulating on the canopy. N is directly absorbed by leaves or in the form of organic compounds. The sample containers were transported to the laboratory in special boxes protecting them from light and heat. The precipitation (mm) amount was measured for each sample, avoiding possible contamination. The samples were pooled for chemical analysis for one sample per plot for each sampling period.

Before chemical analyses, throughfall and precipitation samples were stored at 4 °C. Chemical analysis was performed on throughfall and precipitation samples to determine concentrations of various chemical components, including measurements of pH, sulphates (SO_4^{2-}), nitrates (NO_3^-), chloride (Cl^-), ammonium (NH_4^+), sodium (Na^+), potassium (K^+) and calcium (Ca^{2+}) following the methodology of the ICP Forests monitoring program (Clarke et al. 2016). Samples were prepared by filtration with a membrane filter of 0.45 µm. Concentrations of SO_4^{2-}, NO_3^-, Cl^- and NH_4^+ in throughfall and precipitation were determined using the IC method (Dionex 2010i, Dionex Corporation, USA). Using ion-selective electrodes, Na^+, K^+ and Ca^{2+} concentrations were determined by atomic absorption spectrometry (AAS flame).

Statistical analysis

To calculate the amount of depositions, the total quantity of precipitation received within a specific period (mm per month) was multiplied by the concentration of the compound.

For 2006–2022, we analysed data encompassing monthly precipitation and throughfall amounts, concentrations of ions SO_4^{2-}, SO_4^{2-}–S, NO_3^-, NO_3^-–N, Cl^-, NH_4^+, NH_4^+–N, Na^+, K^+ and Ca^{2+}, as well as depositions of N (sum of NO_3^- and NH_4^+), S, Na, K, and Ca. The dataset used for this study was complete without missing data.

A non-parametric Mann–Kendall test was applied to investigate annual mean concentrations of elements and to determine the monotonic trend over 2006–2022 at different statistical significance levels ($\alpha = 0.001$, 0.01 and 0.05). The Theil-Sen approach was applied to estimate the magnitude of the rate of change per year. The non-parametric Mann–Kendall trend test was used to analyse the climatological and hydrological time series due to its simplicity and robustness (Gavrilov et al. 2016). Moreover, this test identifies monotonic trends in a series of environmental data (Pohlert 2023).

Results

Concentrations of sulphur and nitrogen oxides

Changes in concentrations of air pollutants such as NO_2, NH_3 and SO_2 in the open field next to each forest plot were determined. A significant decrease per decade was found for SO_2 concentrations (Table 2). However, NO_2 and NH_3 concentrations showed uneven results. Since the concentrations of these air pollutants were determined in the open field, the composition of tree species did not affect them. Local meteorological conditions and the varying proximity of the urban centre to the study plots possibly influenced the concentrations, leading to an increase in NO_2 in the mixed coniferous stand. However, a decrease of N concentrations in *P. sylvestris* and *P. abies* stands was identified during the study, although not significant.

Table 2 Average ± standard deviation (SD) of air pollutants and annual trend magnitude, obtained by the nonparametric Mann–Kendall test for each environmental variable

Study plot		NO_2 (μg m^{-3})	NH_3 (μg m^{-3})	SO_2 (μg m^{-3})
Pinus sylvestris (2006–2022)[a]	Average ± SD	10.07 ± 0.52	3.71 ± 0.42	1.61 ± 0.35
	Annual trend magnitude	−0.16	−0.07	−0.11**
Picea abies (2008–2022)	Average ± SD	7.16 ± 0.28	2.68 ± 0.26	1.25 ± 0.12
	Annual trend magnitude	−0.11	−0.06	−0.12*
Pinus sylvestris and *Picea abies* (2006–2017)	Average ± SD	13.91 ± 0.79	3.90 ± 0.27	1.99 ± 0.24
	Annual trend magnitude	0.51	0.15	−0.12

[a]data for the *Pinus sylvestris* stand is available for the 2006–2022, the *Picea abies*—for 2008–2022, and the mixed coniferous stand—for 2006–2017

**$p < 0.001$; **$0.001 < p < 0.01$; *$0.01 < p < 0.05$; $p > 0.05$: not significant

Throughfall and precipitation chemistry

The chemical composition of precipitation is strongly influenced by air pollution, and the presence of a forest canopy introduces additional interactions modifying this composition. The average throughfall pH varied among the sites, ranging from pH 5.7 in the *P. sylvestris* and mixed stands to pH 5.9 in the *P. abies* stand (Table 3). The average over all sites was pH 5.8. Precipitation pH was 0.1–0.3 points higher than throughfall pH, which ranged between pH 5.9 and 6.0.

The average throughfall SO_4^{2-} ranged from 2.5 mg L^{-1} for the *P. sylvestris* stand to 4.9 mg L^{-1} in the mixed stand, with an overall average of 3.7 mg L^{-1} (Table 3). The mean SO_4^{2-} concentrations were 0.3–2.3 mg L^{-1} or up to 1.9 times higher than that in precipitation. Average NO_3^- concentration in throughfall ranged from 5.6 to 10.1 mg L^{-1} for the *P. sylvestris* and mixed stands, respectively. In contrast to the mean NO_3^- concentrations in precipitation, these concentrations were higher, from 1.0 mg L^{-1} in the *P. sylvestris* stand to 2.6–5.8 mg L^{-1} in the *P. abies* and mixed stands. K^+ concentration averages in throughfall ranged from 1.7 to 4.2 mg L^{-1}. On average, they were 2.4–5.6 times higher than that in precipitation.

Mean annual precipitation decreased by − 1.1 mm per decade for throughfall and − 0.6 mm per decade for precipitation for the *P. sylvestris* stand during 2006–2022 (Table 3). Mean annual precipitation for throughfall increased by 1.8 mm per decade for the *P. abies* stand. However, precipitation decreased by − 1.7 mm per decade in the *P. abies* stand during the period. In the mixed *P. sylvestris* and *P. abies* stands, average annual precipitation increased in throughfall (6.0 mm per decade) and precipitation (9.6 mm per decade). There were no significant changes in pH during the study period. In the *P. sylvestris* stand, throughfall depositions of SO_4^{2-} and SO_4^{2-}–S decreased significantly by − 0.11 and − 0.03 mg L^{-1} per decade, respectively. A significant decline was observed in the throughfall depositions of SO_4^{2-} and SO_4^{2-}–S in pure *P. abies* and mixed stands. There was a significant reduction in NH_4^+ and NH_4^+–N concentrations in both throughfall and precipitation across all stand types. There was a significant increase in Ca^{2+} concentrations across all stands. In the *P. sylvestris* stand, Ca^{2+} concentrations in throughfall increased at 0.08 mg L^{-1} per decade, and in precipitation, by 0.12 mg L^{-1} per decade. However, a significant increase was observed only in precipitation for the *P. abies* stand, with an increase of 0.12 mg L^{-1} per decade. A notable increase in Ca^{2+} concentration in precipitation occurred for the mixed stand of 0.16 mg L^{-1} per decade.

Throughfall and precipitation depositions

In *P. abies* (Fig. 2B) and mixed *P. abies* and *P. sylvestris* stands (Fig. 2C), the deposition loads in throughfall represented larger fluxes than in precipitation. Differences between deposition loads show that element fluxes increased by 20–30% when precipitation passed through the canopy. However, there were no differences between the throughfall depositions in the pure *P. sylvestris* stand and precipitation in the open field (Fig. 2A). From the data in Fig. 2, it is apparent that K^+ leached in relatively high quantities from the canopies of both pure coniferous stands. The average K^+ depositions in throughfall were 1.8–4.4 times higher than precipitation.

When comparing the three study plots, the largest difference between element depositions beneath canopies and the open field was in the mixed stand (Fig. 2C). The highest total depositions were identified in the *P. abies* stand (Fig. 2B).

During 2006–2022, mean annual throughfall depositions averaged about 4.1–5.9 kg ha^{-1} of total S, 10.2–25.3 kg ha^{-1} of Cl^-, 6.5–11.8 kg ha^{-1} of Na^+, 8.0–21.7 kg ha^{-1} of K^+, 6.3–7.9 kg ha^{-1} of Ca^{2+}, and 7.9–11.5 kg ha^{-1} of total N (NO_3^-–N + NH_4^+–N) in all stands (Fig. 2).

Annual mean N deposition in throughfall amounted to 7.9 kg ha^{-1} (5.8–10.5) in the *P. sylvestris* stand (Fig. 3A), 8.7 kg ha^{-1} (5.7–15.6) in the *P. abies* stand (Fig. 3B), and 11.5 kg ha^{-1} (9.1–14.1) in the mixed coniferous stand (Fig. 3C). With precipitation, these means were 9.9 kg ha^{-1}, 10.2 kg ha^{-1} and 8.8 kg ha^{-1}, respectively, for stands of *P. sylvestris* and *P. abies* and the mixed stand. In the *P.*

Table 3 Average ± standard deviation (SD) of throughfall and precipitation, and annual trend magnitude obtained by the nonparametric Mann–Kendall test for each environmental variable

Study Plot			Annual precipitation (mm)	Conductivity (μS cm^{-1})	pH	Alkalinity (μeq L^{-1})	SO_4^{2-} (mg L^{-1})	SO_4^{2-}–S (mg L^{-1})	NO_3^- (mg L^{-1})	NO_3^-–N (mg L^{-1})	Cl^- (mg L^{-1})	NH_4^+ (mg L^{-1})	NH_4^+–N (mg L^{-1})	Na^+ (mg L^{-1})	K^+ (mg L^{-1})	Ca^{2+} (mg L^{-1})
Pinus sylvestris (2006–2022)	Throughfall	Average ± SD	558.48 ± 17.17	37.83 ± 1.38	5.68 ± 0.03	3.77 ± 0.45	2.53 ± 0.13	0.88 ± 0.04	5.60 ± 0.35	1.28 ± 0.08	2.02 ± 0.10	0.61 ± 0.05	0.47 ± 0.04	1.19 ± 0.08	1.67 ± 0.12	1.73 ± 0.08
		Annual trend magnitude	−1.08	0.01	0.01	−0.03	−0.11**	−0.03**	0.01	0.02	0	−0.05**	−0.04***	−0.02	0.02	0.08**
	Precipitation	Average ± SD	701.08 ± 26.04	34.01 ± 1.59	5.94 ± 0.04	2.88 ± 0.43	2.28 ± 0.10	0.81 ± 0.03	4.64 ± 0.24	1.14 ± 0.07	1.39 ± 0.08	0.69 ± 0.08	0.54 ± 0.06	1.05 ± 0.06	0.69 ± 0.06	1.97 ± 0.10
		Annual trend magnitude	−0.63	0.44	0	−0.08	−0.03	0	0.13	0.03	0.01	−0.04*	−0.03*	0	0	0.12**
Picea abies (2008–2022)	Throughfall	Average ± SD	582.92 ± 31.44	57.98 ± 1.85	5.90 ± 0.03	2.05 ± 0.34	3.57 ± 0.18	1.22 ± 0.06	6.83 ± 0.34	1.52 ± 0.08	4.49 ± 0.19	0.33 ± 0.04	0.29 ± 0.03	2.11 ± 0.10	3.98 ± 0.20	1.80 ± 0.11
		Annual trend magnitude	1.82	−0.1	0	−0.01	−0.19*	−0.05*	0.03	0.02	0.04	−0.06***	−0.04***	−0.03	0.01	0.06
	Precipitation	Average ± SD	942.00 ± 36.99	34.88 ± 1.81	5.99 ± 0.04	2.50 ± 0.35	2.20 ± 0.10	0.83 ± 0.06	4.21 ± 0.22	1.01 ± 0.06	2.13 ± 0.13	0.37 ± 0.03	0.35 ± 0.04	1.41 ± 0.08	0.71 ± 0.07	2.09 ± 0.13
		Annual trend magnitude	−1.69	0.84	0.01	−0.03	−0.03	−0.02	0.14	0.03	0	−0.04**	−0.03**	−0.04	0.02	0.12*
Pinus sylvestris and *Picea abies* (2006–2017)	Throughfall	Average ± SD	475.05 ± 26.65	66.84 ± 2.86	5.68 ± 0.04	3.88 ± 0.53	4.88 ± 0.34	1.63 ± 0.11	10.12 ± 0.75	2.12 ± 0.15	3.24 ± 0.17	1.52 ± 0.12	1.11 ± 0.09	1.62 ± 0.11	4.17 ± 0.29	1.94 ± 0.15
		Annual trend magnitude	5.95	0.03	−0.03	0.27	−0.40*	−0.08	0.08	0.08	−0.01	−0.16***	−0.10**	−0.07	0	0.13
	Precipitation	Average ± SD	686.54 ± 36.95	33.47 ± 1.70	5.95 ± 0.05	2.86 ± 0.47	2.57 ± 0.13	0.87 ± 0.04	4.28 ± 0.26	0.94 ± 0.06	1.33 ± 0.09	0.70 ± 0.06	0.54 ± 0.05	1.08 ± 0.08	0.74 ± 0.13	1.77 ± 0.12
		Annual trend magnitude	9.56	0.91	0.03	−0.18	−0.04	0.01	0.17	0.03	−0.04	−0.07	−0.06**	−0.04	−0.02	0.16**

***p, < 0.001. **$0.001 < p < 0.01$. *$0.01 < p < 0.05$, $p > 0.05$: not significant

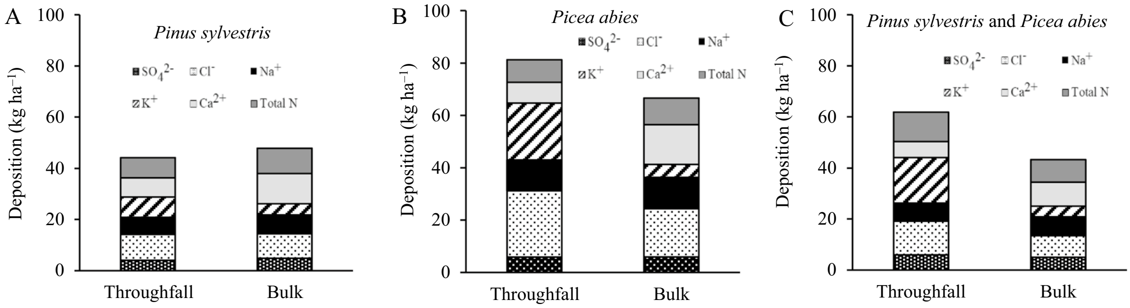
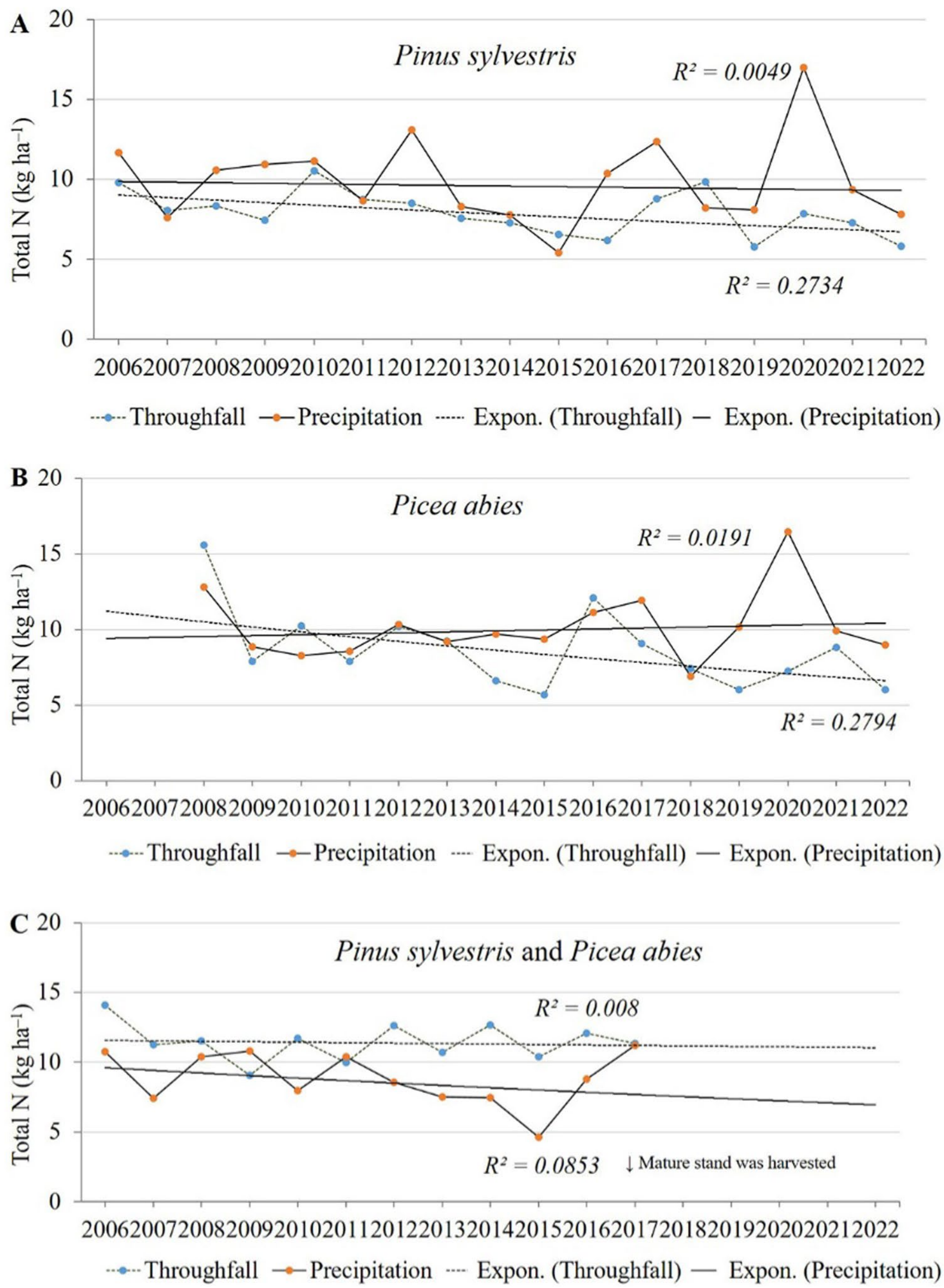


Fig. 2 Annual mean throughfall and depositions (kg ha^{-1}) of total sulphur (S), chloride (Cl^-), sodium (Na^+), potassium (K^+), calcium (Ca^{2+}) and total nitrogen ($N = NH_4^+ + NO_3^-$) in *Pinus sylvestris* (**A**), *Picea abies* (**B**) and mixed *Pinus sylvestris* and *Picea abies* (**C**) stands

Fig. 3 Dynamics of annual deposition of total nitrogen (total $N = NO_3^-{-}N + NH_4^+{-}N$, kg ha^{-1}) with throughfall in *Pinus sylvestris* (**A**), *Picea abies* (**B**) and mixed *Pinus sylvestris* and *Picea abies* (**C**) stands and precipitation over 2006–2022 (missing values: measurements in the *Picea abies* stand started from 2008; measurements in the mixed stand ended in 2017 due to final harvesting)



sylvestris stand, N deposition in precipitation exceeded that in the throughfall by 1.3–1.7 times in 2008–2009, 2012, 2016–1017, 2019 and 2021–2022 (Fig. 3A). In the *P. abies* stand, N deposition in precipitation exceeded deposition in throughfall in 2014–2015, 2017, 2019–2020 and 2022, while no differences were found for the mixed stand.

In throughfall NH_4^+–N depositions, there was an evident decline from 4.3–7.2 to 0.3–2.0 kg ha^{-1} for all stands during 2006–2022 (Fig. 4A, B and C). In contrast, throughfall NO_3^-–N deposition showed either minimal change or an increase throughout the evaluation period (Fig. 4a, b and c). NH_4^+–N and NO_3^-–N depositions in precipitation were more comparable for *P. sylvestris* ($R^2=0.1179$ and 0.3924, respectively) and *P. abies* ($R^2=0.3104$ and 0.3606, respectively) than in the corresponding depositions found beneath canopies.

Potassium depositions showed considerable annual variability, revealing distinct trends in K leaching from tree

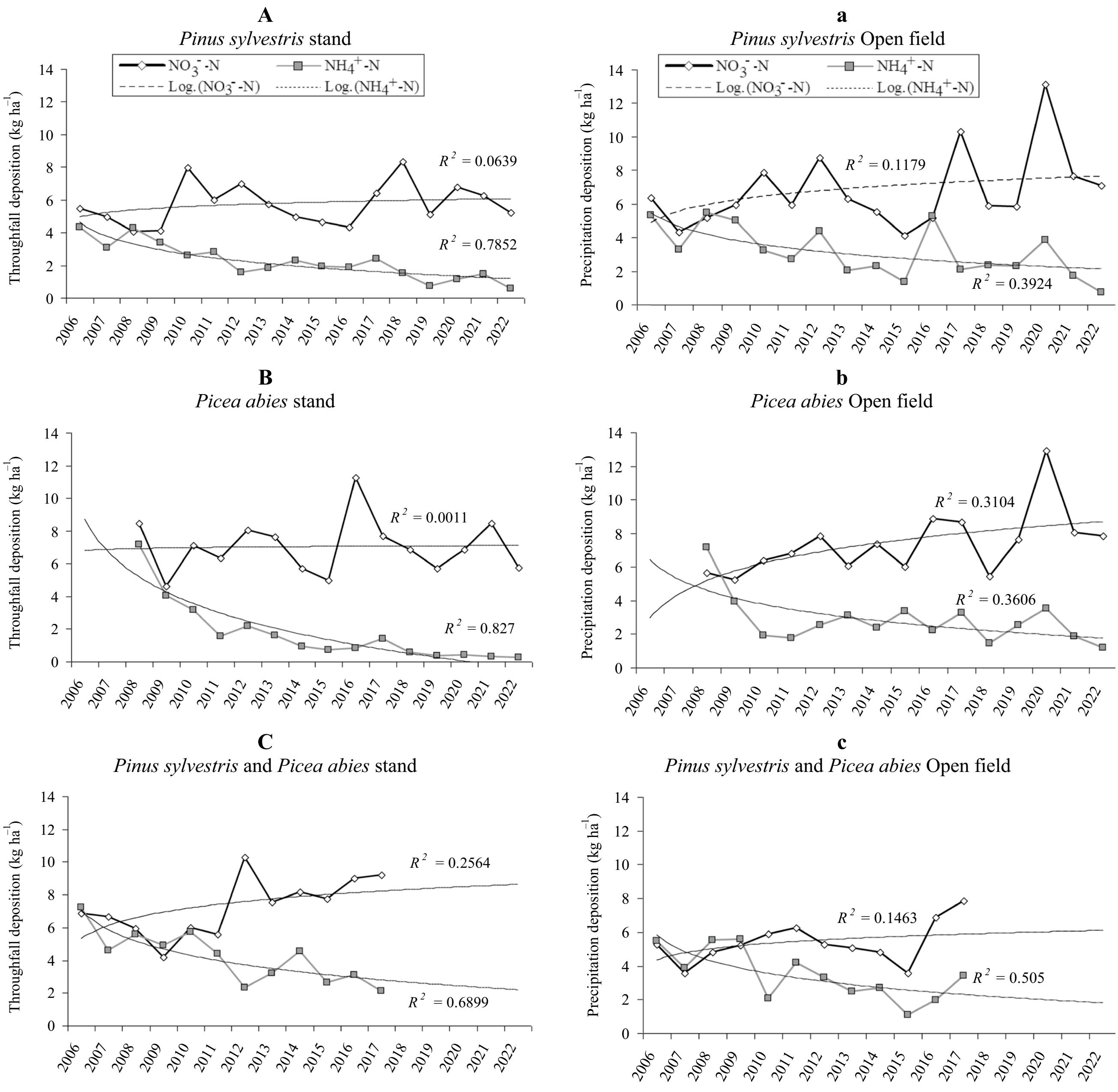


Fig. 4 Dynamics of annual deposition (kg ha^{-1}) of NO_3^-–N and NH_4^+–N with throughfall in *Pinus sylvestris* (**A**), *Picea abies* (**B**) and mixed *Pinus sylvestris* and *Picea abies* (**C**) stands and precipitation in the open field nearby the *Pinus sylvestris* (**a**), *Picea abies* (**b**) and mixed (**c**) stands over 2006–2022

canopies. However, no explicit trends were identified in any stand throughout 2006–2022 (data not shown). Throughfall K depositions ranged from 5.0–11.6 kg ha^{-1} in the *P. sylvestris* stand, 13.4–29.0 kg ha^{-1} in the mixed stand, and 13.1–33.3 kg ha^{-1} in the *P. abies* stand.

Across all plots, S depositions decreased during the study period (Fig. 5).

Despite substantial annual variations in S depositions with precipitation, a distinct decreasing trend was observed in throughfall S depositions (Fig. 5). Between 2006 and 2014, S deposition was 5.3 kg ha^{-1} in the *P. sylvestris* stand, whereas in the period 2015 and 2022, these depositions were nearly half that amount, totalling an average of 2.8 kg ha^{-1} (Fig. 5A). In the *P. abies* stand, throughfall S depositions averaged about 7.4 kg ha^{-1} in 2008–2014 and 4.6 kg ha^{-1} in 2015–2022 (Fig. 5B). Within the mixed stand, these values were 6.7 kg ha^{-1} in 2006–2014 and 3.8 kg ha^{-1} in 2015–2017 (Fig. 5C). The trend of throughfall S depositions

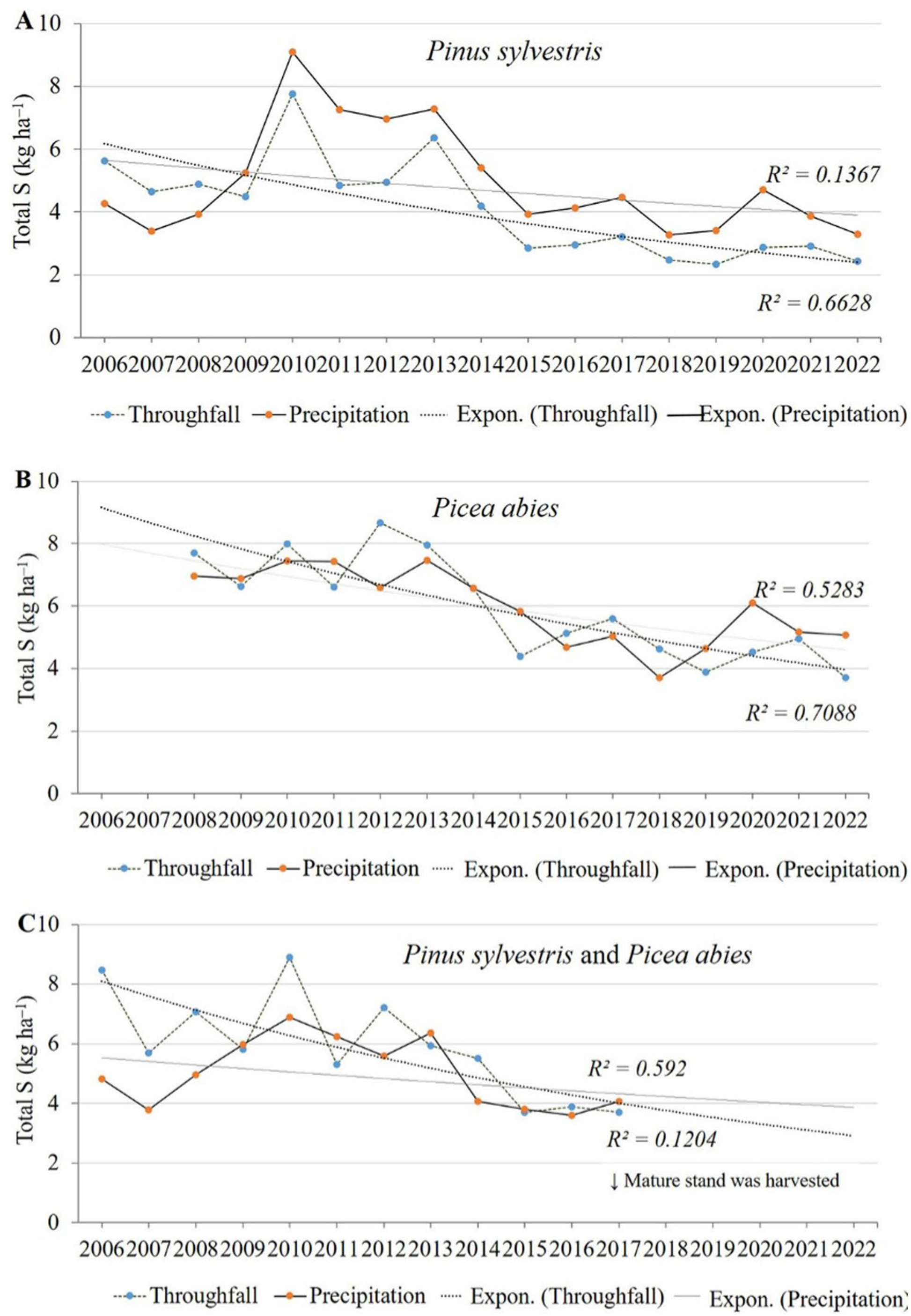


Fig. 5 Dynamics of annual deposition (kg ha^{-1}) of sulphur (S) with throughfall in *Pinus sylvestris* (**A**), *Picea abies* (**B**) and mixed *Pinus sylvestris* and *Picea abies* (**C**) stands and precipitation during 2006–2022

across all three plots showed a decline, with R^2 values ranging between 0.5044 and 0.6691 (data not shown).

Throughfall depositions of Ca^{2+} averaged between 6.3 and 7.9 kg ha^{-1} (data not shown). Significantly, these were 1.5 to 1.9 times lower than those observed in the open field. Over the study period, there was an increase in throughfall depositions of Ca^{2+}, as indicated by the determination coefficient $R^2 = 0.394$ ($y = 0.3403x + 4.4758$) for the *P. sylvestris* stand; $R^2 = 0.490$ ($y = 0.4491x + 4.3348$) for the *P. abies* stand, and $R^2 = 0.569$ ($y = 0.3669x + 3.9144$) for the mixed *stand*.

In the *P. sylvestris* stand, Cl^- depositions were 10.2 ± 1.3 kg ha^{-1} in throughfall and 9.5 ± 1.4 kg ha^{-1} in precipitation; in *P. abies* stands, these depositions were 25.3 ± 1.3 and 18.4 ± 1.3 kg ha^{-1}, respectively, and in the mixed stands, 13.2 ± 0.7 and 8.5 ± 1.9 kg ha^{-1}, respectively (data not shown). Although Cl^- depositions were higher in throughfall than in precipitation in all stands, the highest difference by twice between the values was in the mixed *P. sylvestris* and *P. abies* stand. Over the 2006–2022 period, no significant trend in Cl^- depositions was observed, as indicated by the determination coefficient $R^2 < 0.0905$ for all stands.

Discussion and conclusions

Reports from the UNECE ICP Forests programme indicated a decline in anthropogenic S emissions in Europe over the past several decades (Michel et al. 2022). Nevertheless, there is a significant concern regarding increased N emissions. European forests have experienced a decrease in N and S depositions in recent years (Erisman et al. 2003; Vuorenmaa et al. 2018; Schmitz et al. 2019). This study found a significant decrease in SO_2 concentrations in the ambient air over 2006–2022. Slight decreases were also observed for NO_2 and NH_3 concentrations (Table 2). Chang et al. (2022) assessed long-term changes in precipitation acidity and the presence of acidifying compounds by analysing data from three networks covering North America, Europe, and East Asia. The findings revealed a trend toward rising pH levels in North America and Europe, attributed to decreased SO_2 emissions. In at least these two aspects, the results of the present study showed no significant changes in pH per decade. However, comparable trends were found for changes in SO_2 concentrations in ambient air (Table 2) and for SO_4^{2-} in throughfall and precipitation (Table 3).

Central Europe exhibited high throughfall depositions of NO_3^-–N and SO_4^{2-}–S, while the lowest NO_3^-–N values were detected in Finland and Bulgaria (Michel et al. 2022). Sulphur emissions reduction has decreased the acidity of precipitation, and ammonium compounds have increasingly become more predominant in N deposition (Michel et al. 2022). Over the past few decades, there has been a shift in the NH_4^+–N/NO_3^-–N ratio in atmospheric N, as documented by Liu et al. (2013) and Du (2016). NH_4^+–N/NO_3^-–N ratios have decreased in Lithuania over the 2006–2022 period. This result is like that found for the Netherlands (Boxman et al. 2008). However, this ratio has increased in most European countries (Kurzyca and Frankowski 2017) and in the United States (Du 2016). Changes in NH_4^+–N/NO_3^-–N ratios in ecosystems will most likely lead to changes in plant species richness and productivity (Ren et al. 2021). However, the findings in our study showed relatively low variability in NO_3^- deposition and a clear downward trend in NH_4^+ deposition (Fig. 4).

Another result indicated that canopies of coniferous species decreased the acidity of throughfall, yet led to notably higher concentrations of SO_4^{2-}, NO_3^-, Na^+, and particularly K^+. Total deposition loads increased by 20–30% when precipitation passed through the canopy during 2006–2022 period (Fig. 2). The highest total depositions of the elements were for the *P. abies* stand, while for the *P. sylvestris* stand, differences between the depositions in throughfall and precipitation compared to the open field were low. However, canopies of both conifer species resulted in increased K in throughfall. Eisalou et al. (2013) showed that forest canopy significantly affected pH, total N and P levels, and SO_2, Na and K in the throughfall. Overall K, Ca, Na, and Mg inputs in a forested area were lower than in a non-forested (Tan et al. 2018).

Air pollutants are trapped by tree canopies where primarily N compounds are deposited and washed out by precipitation (Etzold et al. 2020; Ferraretto et al. 2022). Chemical reactions with substances on canopy surfaces, nitrogen oxides ($NO_x = NO_2 + NO$) are converted into less harmful forms. Delaria and Cohen (2023) noted that the deposition of NO_x had significant implications for the oxidative capacity of the regional troposphere, nitrogen inputs in ecosystems, ozone production, and overall air quality. Research has demonstrated that N assimilated by the canopy acts as a nutrient source for broadleaf species, indicating a potential for long-term accumulation within the ecosystem (Da Ros et al. 2023).

Acidification and eutrophication, the main processes driven by nutrient deposition, significantly impact forest ecosystems (de Vries et al. 2015; Costa et al. 2022). Nitrate (NO_3^-) and sulfate (SO_4^{2-}) ions in throughfall are acidifying because they contribute to the acidification of soils and water. However, this study indicated no risk as these ions were unchanged or slightly decreased. This study also found an increase in Ca^{2+}depositions, which suppresses soil acidification. However, as Slootweg et al. (2016) noted, as much as 62% of European ecosystems continued to experience critical eutrophication loads even with the reduction of these quantities.

Research has also demonstrated that nitrogen deposition significantly contributes to the expansion of the carbon sink in forests (de Vries et al. 2014; Du and De Vries 2018; Etzold et al. 2020). This could be considered favourable for climate change mitigation. However, an increased atmospheric N deposition triggers a series of environmental consequences that can adversely affect ecosystems. Changes in soil processes, nutrient imbalances, and changes in vegetation composition are predicted when the annual nitrogen load exceeds 10–20 kg ha^{-1} for deciduous forests and 5–15 kg ha^{-1} for coniferous forests (Bobbink et al. 2011). N depositions, on average from 8.8 to 10.2 kg ha^{-1} in Lithuania during the study period, corresponded to the critical level (Fig. 3).

Essential plant nutrients, such as N, K and Ca, contribute substantially to soil chemistry and are necessary for the growth and development of vegetation. Increased throughfall depositions in forest stands in this study corroborate earlier findings. The ICP Forests Technical Report showed that annual Ca^{2+}depositions were higher than 30 kg ha^{-1} in the Mediterranean and eastern European regions, while in most of northern Europe, these depositions were below 2 kg ha^{-1} (Michel et al. 2022). Annual trends in throughfall depositions, especially K^+, Ca^{2+}, Mg^{2+}, and NH_4^+, were significantly impacted by ion canopy exchange (Nordén 1991; Draaijers and Erisman 1995; Chiwa et al. 2004; Tan et al. 2018).

Despite the reduction of air pollutants and depositions in recent years in Lithuania, fixed depositions were close to or within the range of the critical levels for coniferous forests. Both *P. sylvestris* and *P. abies* stands resulted in a decrease in throughfall acidity and an increase in concentrations of SO_4^{2-}, NO_3^-, Na^+, and particularly K^+. The total amounts of S and N, Cl^-, Na^+, K^+ and Ca^{2+} in the *P. abies* stand exceeded the deposition in the *P. sylvestris* stand (Fig. 2). While K^+ deposition beneath the coniferous canopies exceeded that in the open field due to leaching, leaching of K^+ was considerably lower in the *P. sylvestris* stand compared to the *P. abies* or mixed stands. During the study period, throughfall S depositions over all plots declined. It may be assumed that *P. abies* had a greater effect on pollutant removal than *P. sylvestris*. However, overall, there was no specific effect of tree species on throughfall chemistry, as in all cases, there was an increase in K^+ and Cl^- depositions passing through the canopy. This also agrees with Schwartz et al. (2022), who reported no influence of species composition on throughfall depositions. However, when explaining changes in atmospheric deposition in Lithuania by relating them to local conditions or the specific species composition of forests, it is appropriate to assess the impact of transboundary air pollution, which, due to prevailing western winds, has a strong negative effect on regional environments. While the impacts of increasing nitrogen depositions on forest ecosystems have been studied for several years, future studies should focus on the recovery mechanisms of forest ecosystems as N and S depositions decline (Schmitz et al. 2019). The results provide assumptions about the ability of these coniferous species to remove pollutants not only in forested areas but also in urban areas.

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