Quantification of Enrichment Processes in Throughfall and Stemflow in a Mixed Temperate Forest

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December 4, 2023

Forest ecosystems depend on throughfall and stemflow fluxes for both water and nutrient input. Spatial and temporal variability of throughfall and stemflow fluxes are large and differ between tree species. The nutrient fluxes that accompany throughfall and stemflow are affected by climate, precipitation intensity, the seasonality of dry deposition, and canopy exchange processes. The interdependence of these factors make it challenging to quantify changes in throughfall and stemflow amounts as well as their nutrient content. Here we provide observation-based evidence from 3.5 years of record with 222 rainfall events, of the seasonal variability of throughfall and stemflow magnitude and ion concentrations under a beech *(Fagus silvatica)* and spruce *(Picea abies)* tree. Interception and canopy cover were seasonally variable, average annual interception was 53% below beech, 61% below spruce and 68% below young spruce canopies. Further we assess seasonality of ionic nutrients such as NH_4 and NO_3 as well as Mg, Ca and K and their dependence on both dry deposition and canopy exchange. Throughfall and stemflow were enriched compared to precipitation, with large differences between ions and different months. Antecedent precipitation was a main control on throughfall and stemflow enrichment. We developed a conceptual model of the potential drivers of throughfall and stemflow enrichment based on our observations. While NH_4 and NO_3 enrichment are likely dominated by dry deposition and dew and fog accumulation, Mg, Ca and K were additionally affected by canopy exchange. Observation based studies such as this one are needed to understand precipitation and nutrient partitioning across forests, which enables to predict how changes in climate and forest composition will affect local hydrology and nutrient inputs into forest ecosystems.

1 Introduction

Forests cover 38% of the habitable land area and have major impact on water and nutrient fluxes between the lithosphere and the atmosphere (Betts et al., 2001; Bonan, 2002; Levia and Frost, 2006; Veen et al., 1996). Ongoing climate change will affect the forest ecosystem through changes in precipitation, air temperature and CO_2 concentrations (e.g. Bonan, 2002; Briggs, 2015; *CH2018 - Climate Scenarios for Switzerland; Technical Report* 2018; Nabuurs et al., 2022). Climate change will also impact throughfall and stemflow by which water and nutrients are distributed in the subcanopy and across forests, ultimately affecting forest biogeochemistry (Adriaenssens et al., 2012; Fenn et al., 2013; Kumar Gautam et al., 2017), local stand hydrology and biodiversity, depending on tree species composition (De Schrijver et al., 2007; Levia and Frost, 2006; Nabuurs et al., 2022).

Most water enters the forest subcanopy and forest floor via throughfall (Bren, 2015; Holko et al., 2009; Kofroňová et al., 2021; Krämer and Hölscher, 2009; Levia and Frost, 2006; Mahendrappa, 1990; Mindaš et al., 2018; Ringgaard et al., 2014; Rowe, 1983; Xiao et al., 2000), with lesser contributions from stemflow (Brooks et al., 2012; Draaijers, Van Eak, et al., 1992; Jost et al., 2004; Parker, 1983). This affects root growth of understory vegetation as well as nutrient availability in forest soils (Levia and Frost, 2006; McDowell et al., 2020; Michel et al., 2013; Thimonier, Schmitt, Waldner, and Rihm, 2005). The distribution of throughfall and stemflow is temporally and spatially variable (Levia and Frost, 2006) and affected by precipitation amount (Brooks et al., 2012; Maniak, 1997; McDowell, 1998) and intensity (Brooks et al., 2012), as well as tree type (Staelens, De Schrijver, Verheyen, and Verhoest, 2008), stand density (Macinnis-Ng et al., 2012), and tree characteristics such as canopy cover, branch patterns and stem roughness (Brooks et al., 2012; Macinnis-Ng et al., 2012; McDowell et al., 2020; Staelens, De Schrijver, Verheyen, and Verhoest, 2008). As nutrient input fluxes for the forest ecosystem throughfall and stemflow can on the one hand facilitate diverse ecosystem and on the other hand harm the ecosystem, if certain nutrients become abundant(Eugster and Haeni, 2013; Parker, 1983; Thimonier, Schmitt, Waldner, and Rihm, 2005), It is therefore important to study nutrient inputs on both forest and stand scale (McDowell et al., 2020; Thimonier, Schmitt, Waldner, and Rihm, 2005). Major ion inputs (Cl, Na, Mg, Ca, K, NH_4, NO_3, and NO_2) stem from dry deposition (Andersen and Hovmand, 1999; Liu et al., 2016; Lovett and S. E. Lindberg, 1984; McDowell, 1998; Sun et al., 2014), canopy exchange (Brodo, 1973; Clark et al., 1998; Moffat et al., 2002) and inputs from fog and dew (Groh, Pütz, et al., 2019; Klemm and Wrzesinsky, 2007), which all show distinct seasonality and species dependence (Berger, Inselsbacher, et al., 2009; De Schrijver et al., 2007; Rothe et al., 2002). Although many of these processes are known individually, there still remains a need for observation-driven evidence of the rates and seasonality of nutrient enrichment by throughfall and stemflow in forests.

In this study we investigate climatic forcing of water and nutrient inputs into a forest ecosystem, as well as the species-specific influence of canopy cover on throughfall and stemflow amounts and seasonality. To this end, we compare throughfall and stemflow measurements under beech *(Fagus silvatica)* and spruce *(Picea abies)* to assess the differences between the locally most common deciduous and coniferous tree species in Switzerland.

We collected 222 precipitation, throughfall and stemflow samples at our study site in a mixed temperate forest in Zurich, Switzerland, over the course of 3.5 years. We analysed throughfall and stemflow under a beech, spruce and young spruce canopy each quantitatively and measured the ion concentration in all collected samples to answer the following questions:

- How large is the seasonal variation of throughfall and stemflow under beech and spruce species and to which extent is this variability linked to precipitation intensity and seasonality of canopy cover?
- What are the nutrient enrichment rates of throughfall and stemflow compared to precipitation?
- What are the seasonal dynamics of different nutrient inputs to the forest and which factors explain the seasonality of nutrient enrichment?

2 Study Area and Methods

Our experimental site is a 1.5 km^2 large temperate mixed forest dominated by beech (*Fagus silvatica*) and spruce (*Picea abies*) located at the edge of the city of Zurich (Switzerland), 47°N 8°E, embedded in the larger "Waldlabor Zurich" initiative. The study plot lies in the Holderbach catchment at a mean elevation of 510 m a.s.l. on a hillslope of 20 ° inclination with a mean annual temperature of 9.3 °C and mean annual precipitation of 1134 mm.

Observations from March 2020 until November 2022 are analysed here. During this time, we measured all relevant climate variables just outside the forest with a compact all-in-one weather station (Meter Group - Atmos41) approximately 150 m away from our study site. In the forest, we partitioned our study plot into three subplots: mature beech (B), mature spruce (mS) and young spruce (yS) figure 1a. We measured throughfall (TF) at all three sections and stemflow (SF) at B and mS, as the young spruces at yS do not have stem diameters where stemflow measurements were feasible. Throughfall was measured with 2 m long and 10 cm wide precipitation gutters installed at 1 m distance from the tree stem leading into tipping precipitation gauges with 2 l bottles attached for water sampling. To prevent contamination with organic material, nets were installed at the end of the precipitation gutters to keep larger particles out and to prevent the tipping rain gauges from clogging. Stemflow was measured with a flexible precipitation gutter installed around the tree stem which led into a tipping precipitation gauge and a 2 l bottle for water sampling. We collected samples of precipitation (outside the forest at the meteostation), throughfall and stemflow water, directly after each precipitation event > 3 mm.

To measure canopy cover (CC), i.e. the amount of sky covered by the canopies as seen from the ground, photographs were taken weekly with a DSL camera at the the three plots. Per plot 12 photographs were taken approximately 1 m above ground vertically upwards according to Chianucci (2016) and Chianucci and Cutini (2013) in mode Av (automatic exposure), aperture set to F = 10, a focal length of 55, as well as a exposure correction of -1. From these images the CC was calculated with an automatic threshold function implemented in R following Chianucci and Cutini (2013) with the program in appendix A.1.

Stemflow was measured in volume per time, and in order to compare these measurements with throughfall and precipitation (measured in mm/10 min), the area of the tree canopy needed to be estimated. We followed Hemery et al. (2005) who found a linear relationship between tree diameters at breast height (DBH) > 20 cm and the canopy diameter (DC). For beech the linear fit was determined by Hemery et al. (2005) and Sharma et al. (2017) found values for beech as well as spruce which are reproduced in equations (6) to (7) in appendix A.2. From the canopy diameter the area of the canopy was calculated and measured stemflow volume was transformed to mm/10 min.

Overall we collected samples for 222 precipitation events in the March 2020 to November 2022 observation period, resulting in 607 throughfall samples (n = 210 for B, 198 for mS, 199 for yS) and 380 stemflow samples (209 for B, 171 for mS). All water samples were stored at 4 °C until analysis, filtered using 0.45 μm PTFE Syringe filters (Simplepure, USA) and acidified to a pH between 2 and 3 using 1M HCl. We measured the concentration of major ions Cl^-, Na^+, Mg^{2+}, Ca^{2+}, K^+, $NH_4{}^+$, $NO_3{}^-$, and $NO_2{}^-$, further reported as Cl, Na, Mg, Ca, K, NH_4, NO_3, and NO_2. For all samples we conducted ion chromatography analyses in the Environmental Engineering Laboratory of ETH Zürich (Metrohm Compact IC 761, Metrohm Schweiz AG, Switzerland).

For both beech and mature spruce, event interception (IC) in percent was calculated as the fraction of the measured precipitation outside the forest (PR) which was not reaching the forest floor by throughfall or stemflow, as follows:

$$\mathrm{IC}\ [\%] = 100\,\frac{\mathrm{PR} - \mathrm{TF} - \mathrm{SF}^1}{\mathrm{PR}} \tag{1}$$

[1] for young spruce no stemflow was measured.

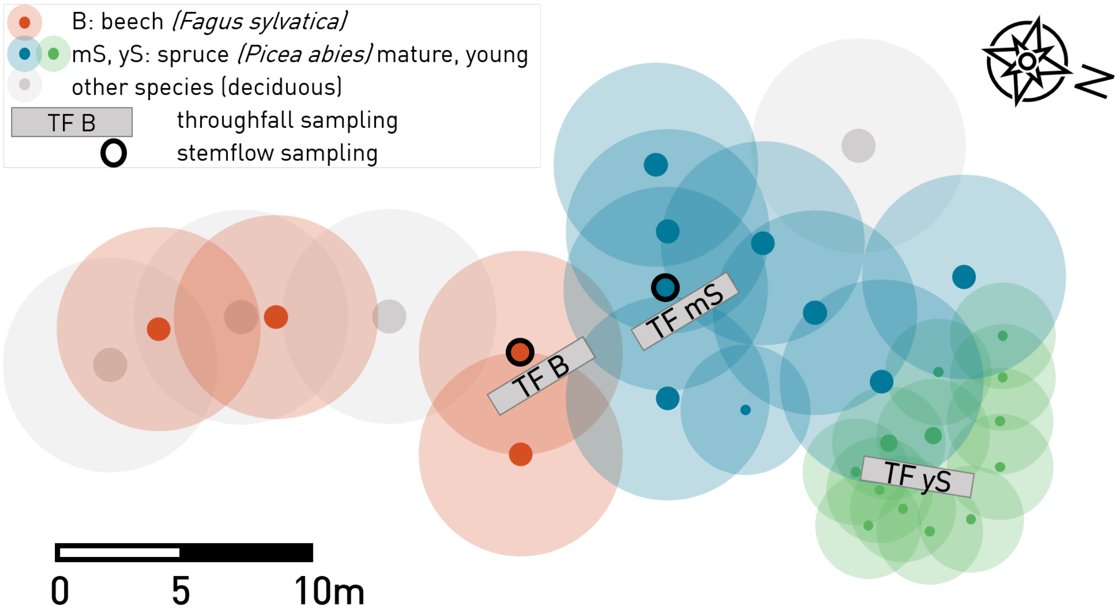
Most results are presented as boxplots, whereas the middle horizontal line of the boxplots indicates the median, the box represents the inter-quartile range, and the whiskers extend to 1.5 times inter-quartile range from the first and third quartiles. The black + signs represent outliers. The numbers in blue below the boxplots indicate the number of samples used to derive the boxplot. We use the Spearman rank correlation to obtain the correlation coefficient rS and p-value statistics to test the significance of the obtained correlations. We use the Wilcoxon-test statistics to analyse significant differences between two groups of samples. We report results as statistically significant when $p < 0.05$.

3 Results and Discussion

3.1 Quantitative Observations of Interception, Throughfall and Stemflow

Interception and stemflow as a function of precipitation per event is shown in Figure 2. Throughfall started after less input precipitation below beech (B) than below mature (mS) and young spruce (yS) trees (mean canopy storage of 1.84 mm for beech and 2.79 mm and 2.84 mm for mature and young spruce, respectively) as indicated by the dashed vertical lines in figure 2a). Eaton et al. (1973) and Maniak (1997) found between 0.5 mm up to 2.5 mm canopy storage for beech and between 1.8 mm up to 4 mm canopy storage for spruce, which are magnitudes comparable to our study results.

Total interception decreased with increasing total precipitation amount as indicated by the logarithmic fit in figure 2a) (see equations (2) to (5) in appendix A.2).



(a) Schematic of our experimental site.

(b) **(c)** **(d)**

Figure 1: Location of "Waldlabor Zürich" study site in Zurich, Switzerland, **(b)** and a schematic of our experimental site at its eastern border **(a)**, indicating the location of single trees (beech, mature and young spruce and other in orange, dark and light green and grey, respectively) and the three subplots (B, mS, yS). The weather station is located outside the forest, approximately 150 m from our experimental site.
Example of two photographs used for canopy cover estimation under mature spruce in early April **(c)** and in late May **(d)**.

Average event interception is biased towards precipitation events which occur more frequently (i.e., small precipitation events). Average per event interception rates (such as shown in figure 2) are 56 % for beech, 57 % for mature spruce and 63 % for young spruce. If precipitation, throughfall and stemflow are integrated over a whole year interception values are 53 % for beech, 61 % for mature spruce and 68 % for young spruce. As interception is measured directly below one respective canopy each, values are not representative for the entire forest stand.

Stemflow amounts are small, with only few events > 1% of total precipitation (as shown in figure 2b)). Beech stemflow showed a mean of 1 % of precipitation and a median of 0.7 %, however for some events there were up to 3.6 % stemflow of total precipitation. Mature spruce showed a mean stemflow of 0.4 %, a median of 0.03 % and a maximum of 3.7 % of total precipitation.

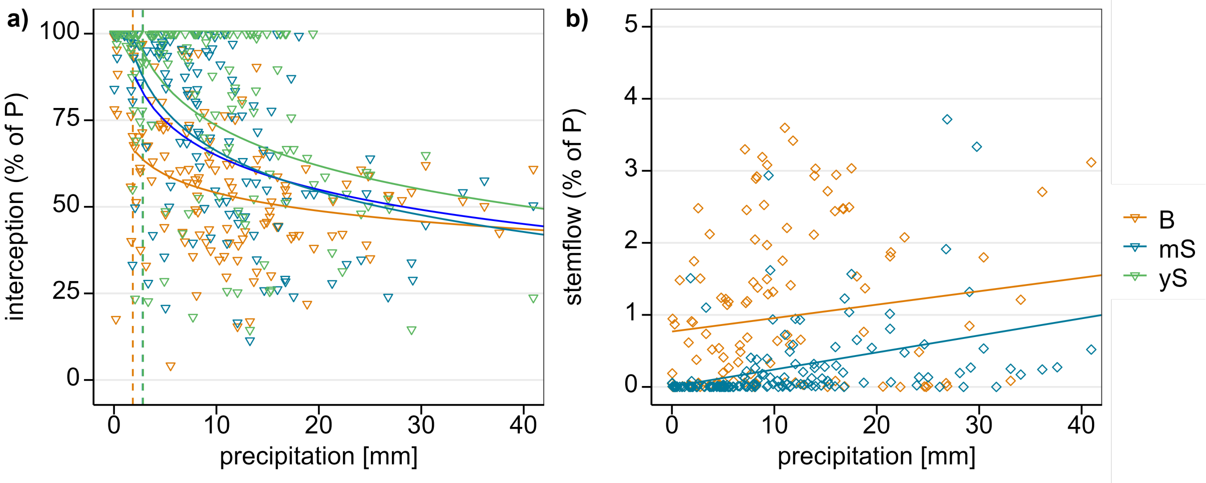


Figure 2: a) Interception as a function of precipitation for beech (B - orange), mature spruce (mS - dark green) and young spruce (yS - light green)). The lines indicate the statistically significant inverse logarithmic fit for beech as well as mature and young spruce for all three species separately and combined (blue line). The vertical dashed lines show the means of the throughfall threshold, which is the mean amount of input precipitation needed to create throughfall, for beech (orange) and mature and young spruce (dark and light green). **b)** Stemflow as a function of precipitation for B and mS. The lines indicate the statistically significant linear fits, respectively.

Interception is therefore mainly affected by throughfall, which in turn is affected by leaf type, leaf and branch surface area, stem roughness and canopy shape (Brooks et al., 2012; Macinnis-Ng et al., 2012; McDowell et al., 2020) as well as intensity and distribution of precipitation (Brooks et al., 2012) and thus shows high spatial and temporal variability (Levia and Frost, 2006). Before reaching the soil the forest-floor litter layer further intercepts precipitation (Brooks et al., 2012; Floriancic et al., 2022).

Previous studies found stemflow to be between 1 and 10 % of annual precipitation (Bren, 2015; Brooks et al., 2012; Draaijers, Van Eak, et al., 1992; Jost et al., 2004). Stemflow under deciduous trees is generally larger than under coniferous trees (Levia, Keim, et al., 2011; Ponette-González et al., 2020; Van Stan and Stubbins, 2018), being affected by branch altitude, the shape of crown or canopy structure and the roughness of bark (Brooks et al., 2012; M. S. Johnson and Lehmann, 2006; Levia, Nanko, et al., 2019). Beech having smoother bark and a more funnel shaped canopy than spruce could therefore contribute to the differences in stemflow. Overall, we found that stemflow volume is negligible, but differs between beech and spruce and should be evaluated for different tree species separately. The possible relevance as input for nutrients is discussed in section 3.3.1.

The fraction of interception decreases with increasing precipitation event amount and can be described with an asymptotic decrease of interception with increasing precipitation, similar to what we show in figure 2a) and equations (2) to (5) (Darryl E. Carlyle-Moses and Gash, 2011; Maniak, 1997; McDowell et al., 2020). For small precipitation events measuring throughfall becomes increasingly difficult, an issue which can be mitigated by using more rain gauges or larger throughfall sampling areas (Cuartas et al., 2007; Price and D. E. Carlyle-Moses, 2003). Literature values for canopy interception range from 9 % to 29 % of annual precipitation for beech forests (Bren, 2015; Krämer and Hölscher, 2009; Mindaš et al., 2018; Rowe, 1983) and from 21 % to 37 % for spruce forests (Holko et al., 2009; Kofroňová et al., 2021; Mahendrappa, 1990; Ringgaard et al., 2014; Xiao et al., 2000). For individual events Puncochar et al. (2012) reported interception values of 44 % up to 65 % for a predominantly coniferous forest. Some studies show higher interception at deciduous stands compared to coniferous stands, which is the opposite of what we found in our data (Darryl E. Carlyle-Moses and Gash, 2011; Snakin et al., 2001; Thimonier, Schmitt, Waldner, and Rihm, 2005). The generalising grouping into coniferous and deciduous trees could be misleading, and more objective parameters such as leaf area, canopy cover or stand density should be assessed for interception comparisons.

Uncertainties in these measurements stem from the large variability of both precipitation and throughfall, possibly enhanced by the proximity of our site to the forest edge, and the limited measurements under only one tree canopy for each species.

3.2 Seasonality of Canopy Cover and Interception

The seasonality of canopy cover and interception for the three plots beech, mature spruce and young spruce are shown in figure 3. Example pictures of how canopy cover was assessed from pictures taken with a DCLR camera are shown in figures 1c to 1d. While beech showed a clear, statistically significant seasonality with lower canopy cover in winter (DJF) and spring (MAM) and higher canopy cover in summer (JJA) and autumn (SON), the canopy cover for the spruce trees was constantly high across the entire year. At our study site, canopy cover under beech increased from day 115 of the year on (28th of April) and decreases from day 285 on, which spans the typical growing season of beech (Ahrends et al., 2008; Prislan et al., 2019; Yang et al., 2017). Canopy cover is an easily measurable proxy for leaf area index (Chianucci and Cutini, 2013), which influences the amount and the enrichment of throughfall and stemflow (Draaijers, Van Eak, et al., 1992; McDowell et al., 2020). Interception at our site showed less distinct seasonal differences (figure 3b)) than canopy cover. Some seasonality of interception was evident for beech, however much less pronounced than the canopy-cover seasonality. Interception decreases from summer (JJA) to winter (DJF) (56 % to 54 % for B, 88 % to 61 % for mS and 100 % to 64 % for yS). As changes in canopy cover cannot explain these changes, other factors such as precipitation intensity may play a role. Mean precipitation intensities had a strong seasonal variability (figure 3) with low intensity precipitation being dominant in

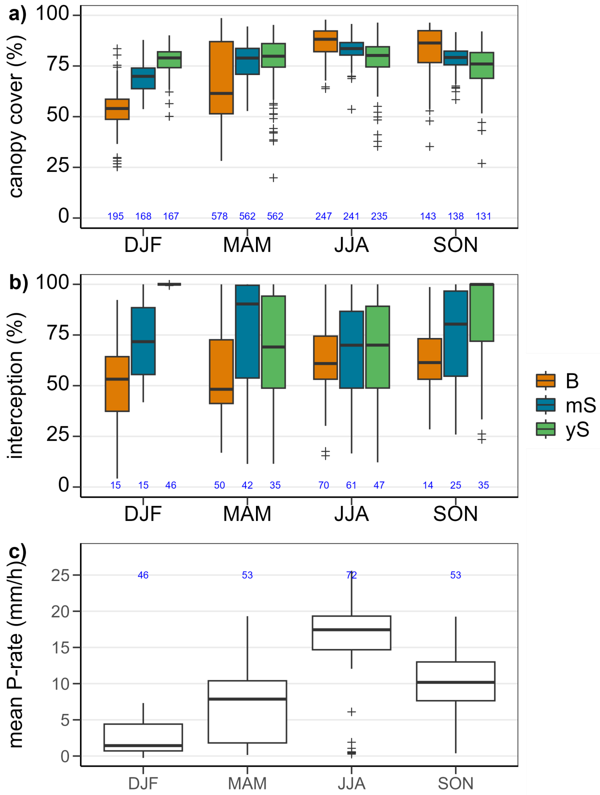


Figure 3: Canopy cover **(a)** % and interception **(b)** % in winter (DJF), spring (MAM), summer (JJA) and autumn (SON) below beech (orange), mature spruce (dark green) and young spruce (light green) as well as the maximum precipitation intensity **(c)** as a mean for each event in mm/h.

winter. We found median precipitation intensities of 0.24 mm/h in winter and 0.42 mm/h in summer, which is an indicator of more convective precipitation in summer and more stratiform precipitation in winter.

Higher canopy cover increases interception (Staelens, De Schrijver, Verheyen, and Verhoest, 2008). In our study throughfall was generally lower in the dormant season (figure 3) under both beech and spruce, even though canopy cover was larger in summer for the beech canopy. As spruce canopy cover was almost constant over the year, the lower interception in spruce canopies in winter is the result of other factors, such as precipitation patterns like precipitation intensity, which can be seen in figure 10a). The seasonal development of the interception under spruce can be interpreted as solely a function of factors such as precipitation intensity and amount, while beech seasonal interception development is additionally affected by the seasonality of the canopy cover.

3.3 Enrichment processes

Nutrient enrichment processes in the forest are mainly affected by precipitation and its origin, but also by factors such as geographic location and surrounding geology, tree type, climate and weather, dry deposition, canopy exchange from leaves, needles and branches, and uptake and release from epiphytic organisms such as lichens, mosses, and algae (Akkoyunlu and Tayanç, 2003; McDowell et al., 2020; Novak et al., 2020; Polkowska et al., 2005). In the following section we will show and qualitatively and quantitatively discuss the enrichment processes below beech and spruce in section 3.3.1, after long dry periods in section 3.3.2, as well as the seasonality on enrichment in section 3.4, and the seasonal enrichment of salt, nitrogen and geogenic ions specifically in sections 3.4.1 to 3.4.3. Further we will discuss potential forcing of seasonal enrichment in section 3.5.

Enrichment in throughfall and stemflow will be displayed as the ion concentration measured in throughfall or stemflow minus the ion concentration measured in precipitation for the major ions Cl, Na, Mg, Ca, K, NH_4, NO_3 and NO_2.

3.3.1 Enrichment processes in beech and spruce canopies

Figure 4a) shows the enrichment in throughfall and stemflow relative to precipitation for the major ions Cl, Na, Mg, Ca, K, NH_4, NO_3 and NO_2. We measured median enrichment of up to 0.15 mmol/l in throughfall and 0.18 mmol/l in stemflow, whereas K enrichment was strongest for both throughfall and stemflow. Differences in enrichment between stemflow and throughfall were statistically significant for Ca, K and NH_4. In figure 4b) and c) we show the enrichment for all ions for beech, mature and young spruce for throughfall and stemflow, respectively. Enrichment was largest for Ca, K and NH_4 ions. Differences in throughfall between beech and mature spruce were significant for all ion species except for Ca and NO_2. Differences in throughfall between beech and young spruce were significant for all ion species except for Ca and NO_2. Differences in throughfall between mature and young spruce were significant for Na only. Differences in stemflow between beech and mature spruce were significant for all ion species except for NO_2.

Enrichment in stemflow is significantly larger than in throughfall for only Ca, K and NH_4 (figure 4). Understanding the processes that affect throughfall and stemflow differently are difficult to distinguish, as water in both, throughfall and stemflow, may come in contact with several layers of both leaves or needles, branches and stems (McDowell et al., 2020; Parker, 1983). For a conceptual understanding, throughfall and stemflow should possibly rather be viewed on a spectrum of longer or shorter contact with either leaves, branches or the stem. More contact would lead to stronger enrichment but also greater interception, which would explain both the larger beech stemflow amounts as well as the higher concentrations of spruce stemflow.

Whereas throughfall affects a larger area of the forest floor and is volumetrically prominent, stemflow can affect the stand scale nutrient availability (Chang and Matzner, 2000; Parker, 1983; Thimonier, Schmitt, Waldner, and Rihm, 2005). The contribution of stemflow to the overall ion concentration a forest stand receives might therefore be higher than its contribution to the hydrological fluxes (Neary and Gizyn, 1994). Some link larger stemflow nutrient input to larger stemflow quantities, while others expect bark ionic composition of affect the nutrient content of stemflow (Adriaenssens et al., 2012; Parker, 1983). Nitrogen was found to be more enriched under spruce than under beech, possibly due to dry deposition on the rough bark and branches of spruce compared to beech (Berger, Inselsbacher, et al., 2009; De Schrijver et al., 2007).

The single effects of canopy shape, properties of leaves, branches and stem and tree interaction could not be separated in the scope of this study and remain to be looked at in further research. Overall, our results suggest that differentiating between beech

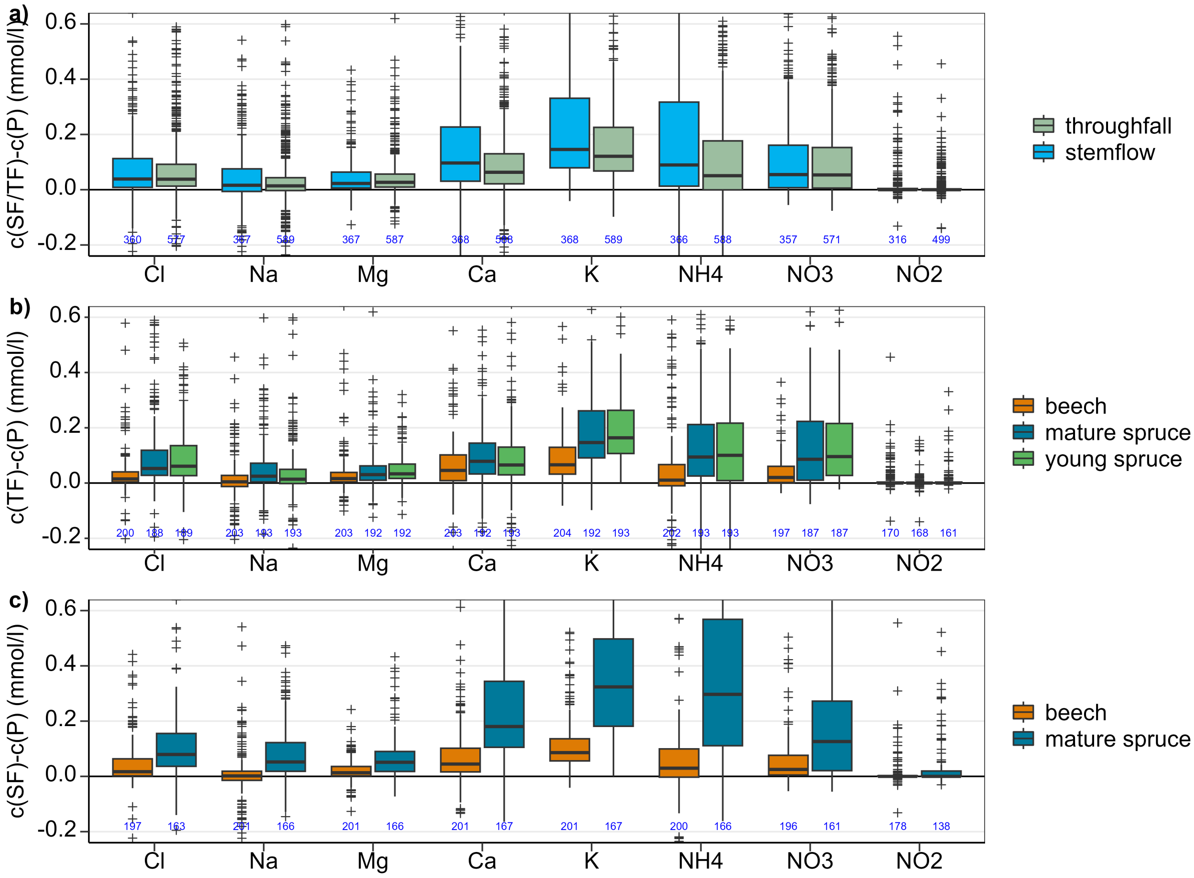


Figure 4: a) Enrichment in throughfall (blue boxplots) and stemflow (green boxplots) for all tree species together, **b)** enrichment in throughfall for beech (orange), mature spruce (dark green) and young spruce (light green), **c)** enrichment in stemflow for beech, major and young spruce.

and spruce increases the understanding of both volume (see section 3.1) as well as ion concentration in throughfall and stemflow. Both throughfall and stemflow get enriched in magnitudes larger than the concentrations found in precipitation (see figure 5a)).

3.3.2 Enrichment processes in relation to antecedent precipitation

We tested the hypothesis that fewer antecedent precipitation leads to stronger enrichment signals in throughfall and stemflow (Berger, Untersteiner, et al., 2008; McDowell, 1998). This would be the case when rainfall after a long dry period would flush dry deposition off the canopy. Therefore, we divided the precipitation event data into four quartiles depending on the amount of precipitation in the 10 days prior to the precipitation event, and plotted the measured enrichment in figure 5. Plot **a)** shows the concentrations measured in precipitation, **b)** shows the enrichment in throughfall and **c)** shows the enrichment in stemflow for all measured ions. We performed a Mann-Kendall-test on the data ordered by the amount of antecedent precipitation, to assess the significance of the trend that drier antecedent conditions lead to larger enrichment. For precipitation, the trend was negative and significant for Cl, K and NH_4. For throughfall the ion concentrations for an increasing amount of antecedent precipitation showed a significant negative trend for the following ions: Cl>Mg>Ca>K>NH_4>Na>NO_3 (in order of the magnitude of the trend). For stemflow, the ion concentrations for an increasing amount of antecedent precipitation showed a significant negative trend for the following ions: Cl>Mg>NH_4>K>Ca>NO_3 (in order of the magnitude of the trend).

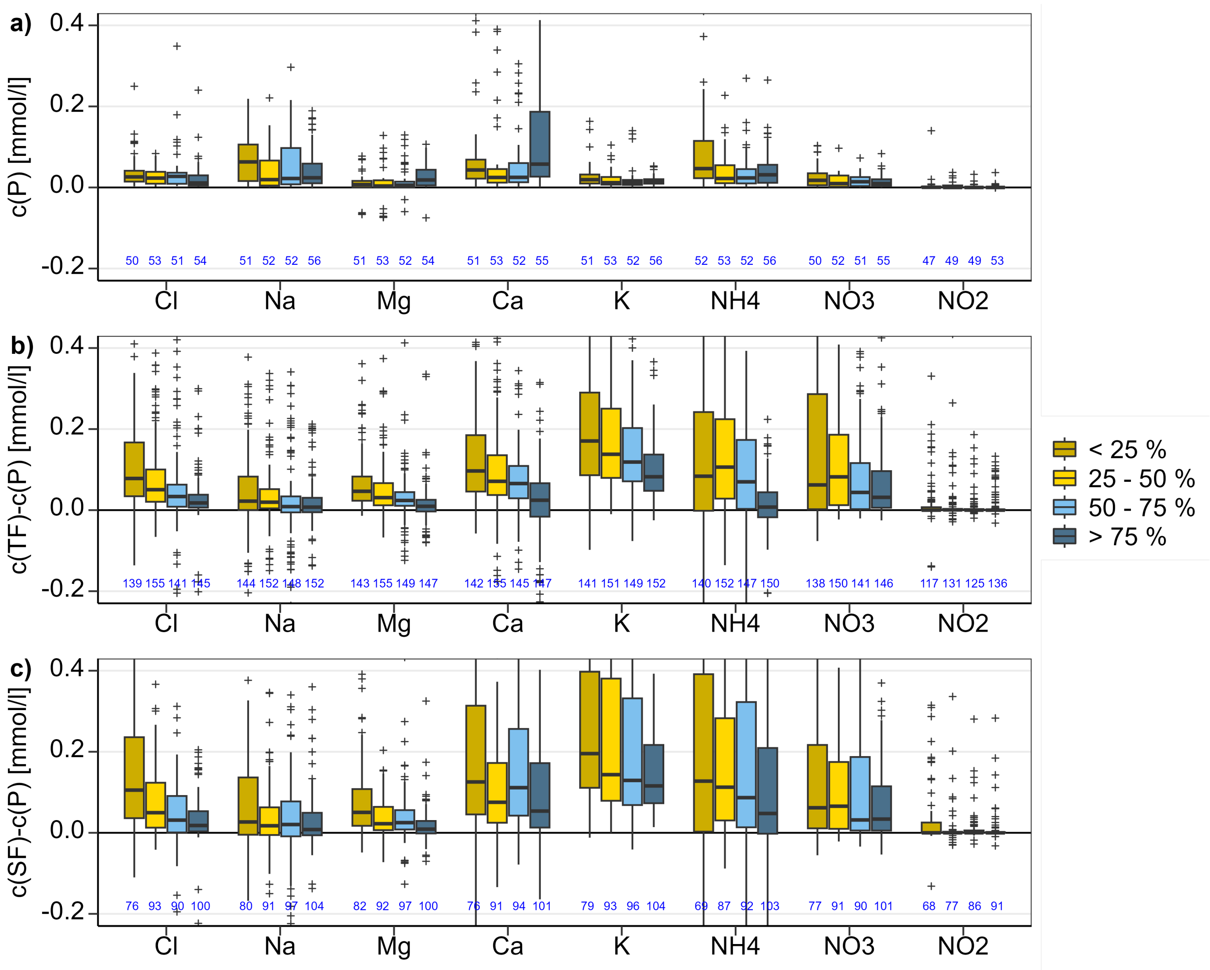


Figure 5: Ion concentrations in **a)** precipitation, **b)** throughfall and **c)** stemflow for the four quartiles of antecedent precipitation in the 10-day periods prior sampling of our record for the major ions Cl, Na, Mg, Ca, K, NH_4, NO_3 and NO_2. The blue numbers indicate the absolute numbers of cases that shape the boxplots.

We see significantly higher ion concentrations in throughfall and stemflow after longer periods of no precipitation, which was also observed by Berger, Untersteiner, et al. (2008) and McDowell (1998). The difference between precipitation and throughfall and stemflow which we show in figure 5b) and c) can be seen as a measurement of dry deposition of nutrients, if we assume no or negligible exchange processes by the tree (Berger, Untersteiner, et al., 2008; Staelens, Schrijver, et al., 2005; Thimonier, Schmitt, Waldner, and Rihm, 2005). Dry deposition happens as aerosol particles settle on canopy leaves, branches or stems, and is larger in forests is larger than in the open field, and stronger at the forest edge than in its centre (Adriaenssens et al., 2012; Draaijers, Van Eak, et al., 1992; McDowell et al., 2020; Thimonier, Schmitt, Waldner, and Rihm, 2005).

The observed dry deposition is most likely a relevant nutrient input into the forest, and the observed magnitude shows the relevance of dry deposition at our study location.

3.4 Seasonal patterns of enrichment processes

We found seasonal patterns in ion concentration in precipitation, throughfall and stemflow, as shown in figure 6a), b) and c), respectively. The seasonal patterns were stronger in throughfall and stemflow than in precipitation. Enrichment was strongest in spring (MAM), followed by autumn (SON) and summer (JJA) with the weakest enrichment found in winter (DJF) when averaged across all measured ion species. Seasonality of the salt ions Cl and Na is further discussed in section 3.4.1, the seasonality of geogenic ions Ca, K, Mg in section 3.4.2 and the seasonality of nitrogen species NH_4, NO_3 and NO_2 in section 3.4.3.

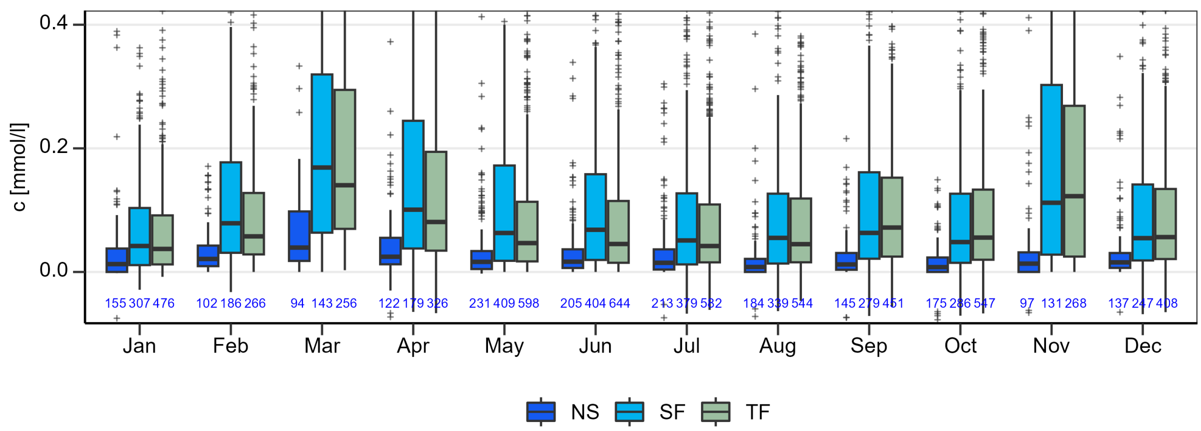


Figure 6: Seasonal variation in ion concentrations of all ions for precipitation (dark blue) as well as throughfall (green) and stemflow (blue). The blue numbers indicate the total number of samples used to compile the boxplots.

Seasonality of ion enrichment is reported to be primarily a function of climate seasonality, being dependent on precipitation variance, presence or absence of canopy cover and differences in dry deposition, exchange or dew and fog (Akkoyunlu and Tayanç, 2003; Berger, Untersteiner, et al., 2008; Draaijers, Van Eak, et al., 1992; Groh, Slawitsch, et al., 2018; Klemm and Wrzesinsky, 2007; Levia and Frost, 2006; McDowell, 1998; Moffat et al., 2002).

3.4.1 Salt

Concentrations of Na and Cl and their difference in precipitation (figure 7a)) are low for most of the year with a slight increase from February until April. For precipitation there is no significance difference between the concentrations of Na and Cl. In throughfall and stemflow the concentrations of both Na and Cl are enriched with peak enrichment taking place from February until May for both Na and Cl as shown in figure 7b) and c), respectively. Cl shows an additional peak in enrichment in November. Concentrations reach their yearly low from June until August. The difference of the concentrations in Na and Cl are plotted in all three panels of figure 7 to show disconnection of the concentrations in Na and Cl.

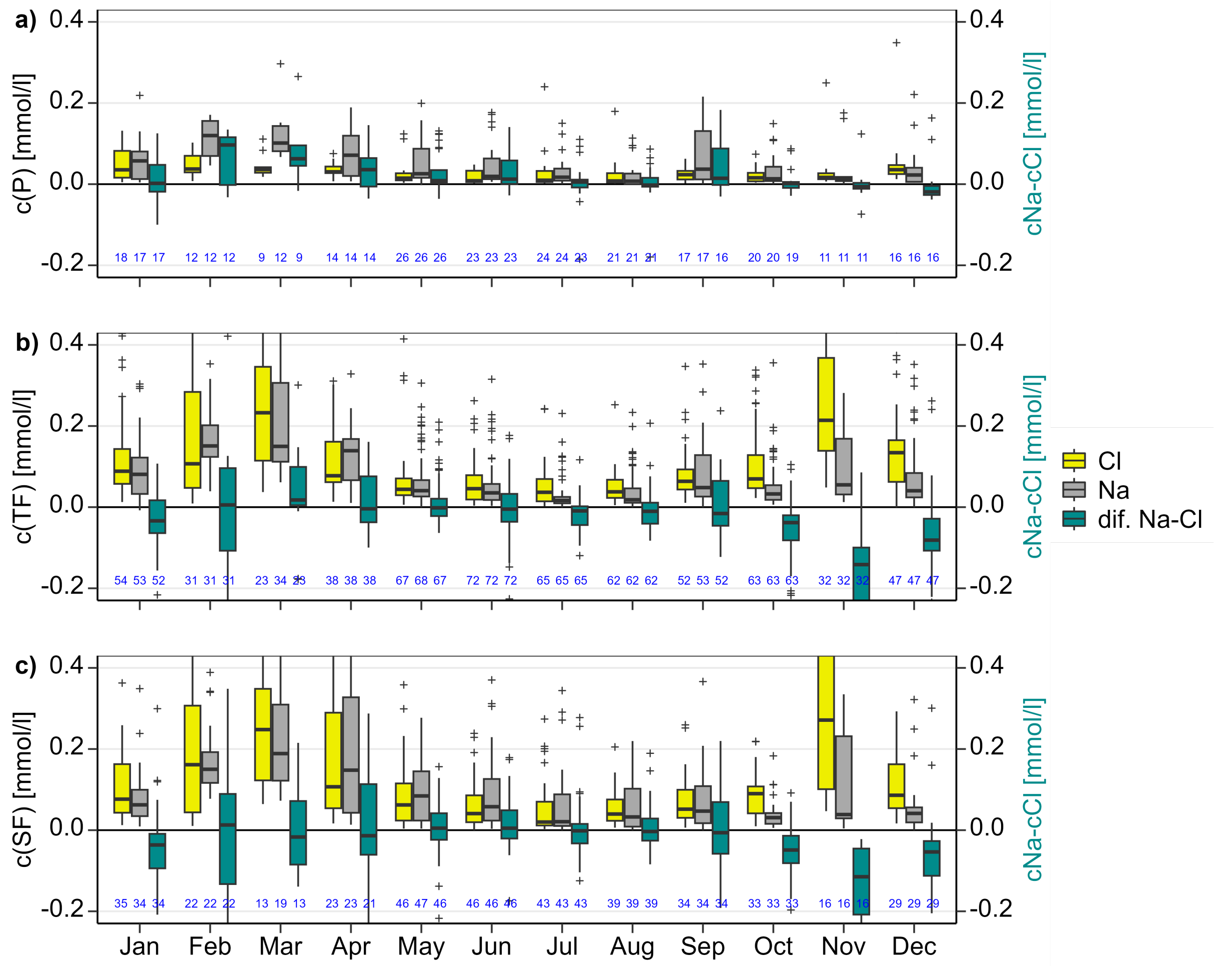


Figure 7: Seasonal variability of the concentration of Cl (yellow) and Na (gray) as well as the difference in Cl and Na concentration (in cyan) in **a)** precipitation, **b)** throughfall and **c)** stemflow. The blue numbers indicate the total number of samples used to compile the boxplots.

As Na and Cl are relatively inert, they can be used as a proxy for dry deposition, which in our case might be amplified by the forest edge (Draaijers, Van Eak, et al., 1992; McDowell et al., 2020). The high concentrations in spring and autumn point to the impact of maritime sources of the low pressure systems crossing middle Europe in spring and autumn from the west, (Moffat et al., 2002; Thimonier, Schmitt, Waldner, and Schleppi, 2008). The molecular formula of sodium-chloride salt is NaCl, meaning that if coming from the crystalline form, as would be expected in dry deposition, the Na:Cl ratio should be 1:1. As evident from the large molar discrepancy seen in figure 7b) and c) from October until January, there must be another source of Cl or a sink of Na which leads to this disconnection of the concentrations. Na is often used as a tracer ion as it is only deposited in particles and canopy exchange rates for Na are low (Macinnis-Ng et al., 2012; Staelens, De Schrijver, Verheyen, and Verhoest, 2008).

A possible explanation for higher Cl concentrations would be salt brime used to de-ice roads during the winter months. (Thimonier, Schmitt, Waldner, and Rihm, 2005). As no temporally resolved data on the salt use in the study area exists, we can only compare our data to annual data: According to the salt use of the Canton of Zurich (*Kanton Zürich; Strassennetz; Winterdienst; Salzverbrauch* 2022), responsible for the de-icing of the closest road to the study site, in the winter of 2020/21 10 times more salt was used than in the winter of 2021/22 and we observed 2-times higher salt concentrations in the 2021/22 winter compared to the winter 2020/21.

3.4.2 Geogenic ions

The ion concentrations of Mg, Ca and K are shown in figure 8 for precipitation, throughfall and stemflow.

The concentrations in precipitation were low or below detection limit all year round. For throughfall and stemflow however, we observed an annual pattern of highest concentrations in November, high concentrations in spring and summer and lowest concentrations in December, January and February.

Mg had its concentration peaks in throughfall and stemflow in April and May and had significantly higher concentrations after long dry periods (figure 5).

Ca concentrations in throughfall and stemflow were relatively homogeneous all year round with lowest concentrations during December and January and highest concentrations in November, where the median was two times higher than during the rest of the year. Ca showed high differences between beech and spruce for stemflow (figure 4c)), having significantly higher concentrations in stemflow than in throughfall. Events after long dry periods (figure 5) had significantly higher concentrations.

K had its highest concentration in throughfall and stemflow in November, followed by its lowest concentrations in December, January and February with an increase in concentration again in March and April and a decrease across the summer months. K concentrations were higher in spruce stemflow than in beech stemflow (figure 4c)) and they were significantly higher in stemflow than in throughfall. Longer dry periods had a significant effect on K concentration in both throughfall and stemflow (figure 5).

Mg, Ca and K mainly originate from rock weathering and are therefore highly dependent on geographical location (Botter et al., 2019; Polkowska et al., 2005). Kumar Gautam et al. (2017) reported enrichment in Ca and Mg stemming from dry deposition, with Ca having the highest dry deposition and K the lowest, which stands in contrast to our finding of much higher enrichment in K and Ca than in Mg. Reasons for that might be differences in the surrounding bedrock. As our data does not show the same seasonality as the biologically less active Na and Cl ions (figure 7), we assume that there are other sources of Mg, Ca and K, such as leaching processes from the canopy. Thus, it is likely that peaks in Mg, Ca and K at our site did not originate from dry deposition, but rather from canopy leaching. Mg concentrations were similar in beech and in spruce in both throughfall and stemflow (figure 4b) and c)), which is a further indicator that the Mg concentrations did not originate from dry deposition, which generally affected concentrations under spruce much more than under beech. According to D. W. Johnson and Steven E. Lindberg (1992) and Parker (1983) Mg, Ca and K are getting leached from the canopy, which might be increased by acid precipitation (Polkowska et al., 2005). Variations however are being reported in the amount of leaching, whereas Rothe et al. (2002) reported higher leaching in Mg than in Ca, which is not what we found at our study site.

Levia and Frost (2006) reported an increase in Ca concentration when the amount of throughfall decreased. This connection between decreasing amounts of throughfall and

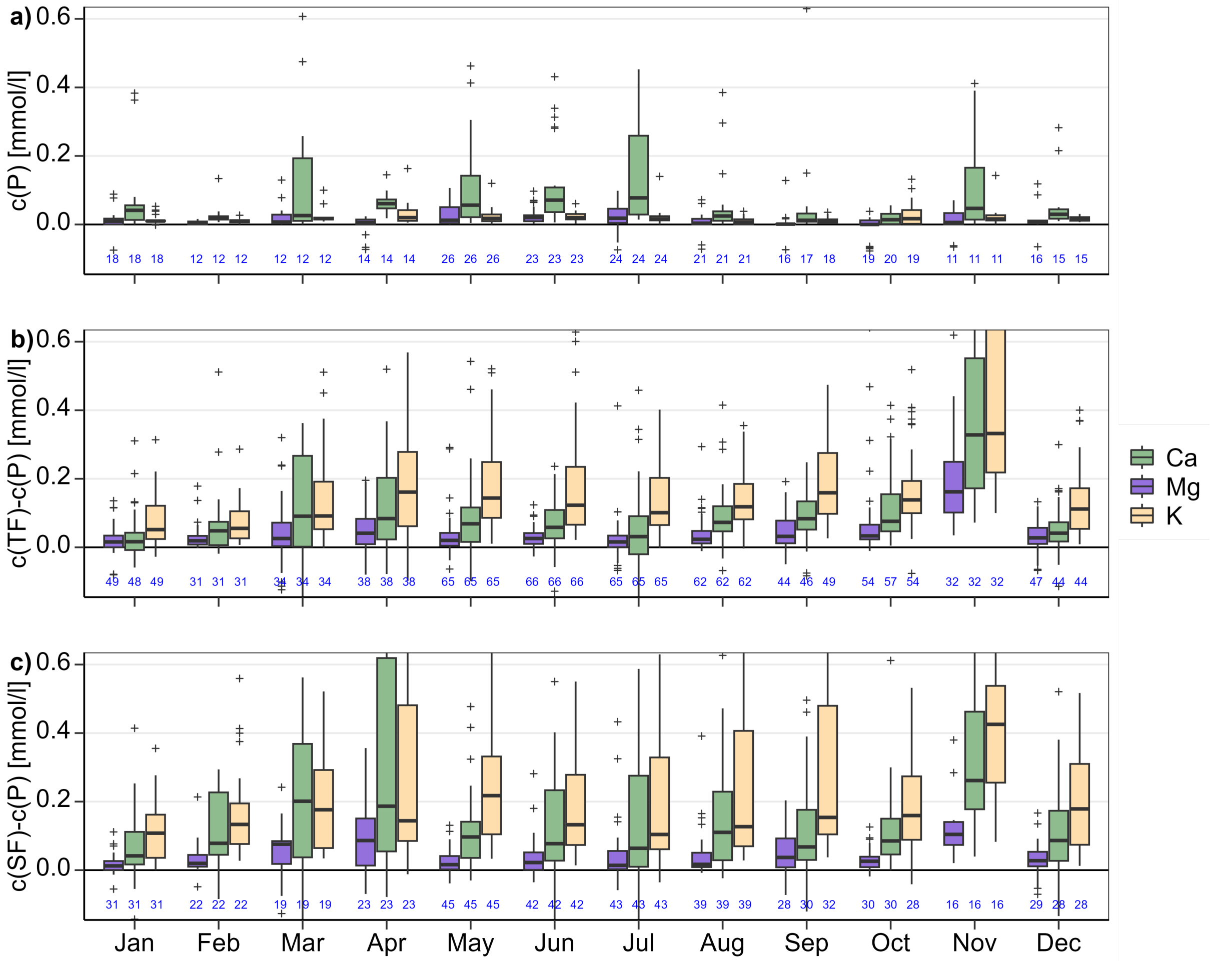


Figure 8: Seasonal variability of the concentrations of calcium [Ca] (violet), magnesium [Mg] (green) and potassium [K] (beige) in **a)** precipitation, **b)** throughfall and **c)** stemflow. The blue numbers indicate the total number of samples used to compile the boxplots.

increasing Ca concentrations is also evident from our data figure 5b).

Concluding, the geogenic ions (Mg, Ca, K) showed a different seasonality than other ions, which suggests that leaching, in combination with dry deposition, played a more important role for these ions at our site. This could potentially explain peaks in November, when wind is more abundant at our site. If this is correct, the concentrations in stemflow should be larger than in throughfall, which is only true for K and not for Mg and Ca, so there might be some leaching or another process involved.

3.4.3 Nitrogen

Nitrogen was plotted in NH_4, NO_3 and NO_2 for precipitation, throughfall and stemflow in figure 9. The concentration in precipitation of NO_2 was just around detection limit for almost all measurements, and the enrichment compared to precipitation was zero for most months for both throughfall and stemflow, with the exception of an increase in NO_2 concentrations in April in both throughfall and stemflow.

Concentrations of NH_4 in precipitation and to a lesser extent also NO_3 only increased in March and April (figure 9a)).

For throughfall and stemflow we found a strong seasonal pattern with strongest concentrations in spring, followed by autumn, summer and winter, with peaks in concentrations in March and September for both NH_4 and NO_3. NH_4 and NO_3 showed different enrichment patterns in throughfall and stemflow, the latter had two times the concentrations of throughfall from May until November for NH_4. Enrichment in stemflow was much more variable than in throughfall, especially for NH_4 and enrichment was strongest in summer.

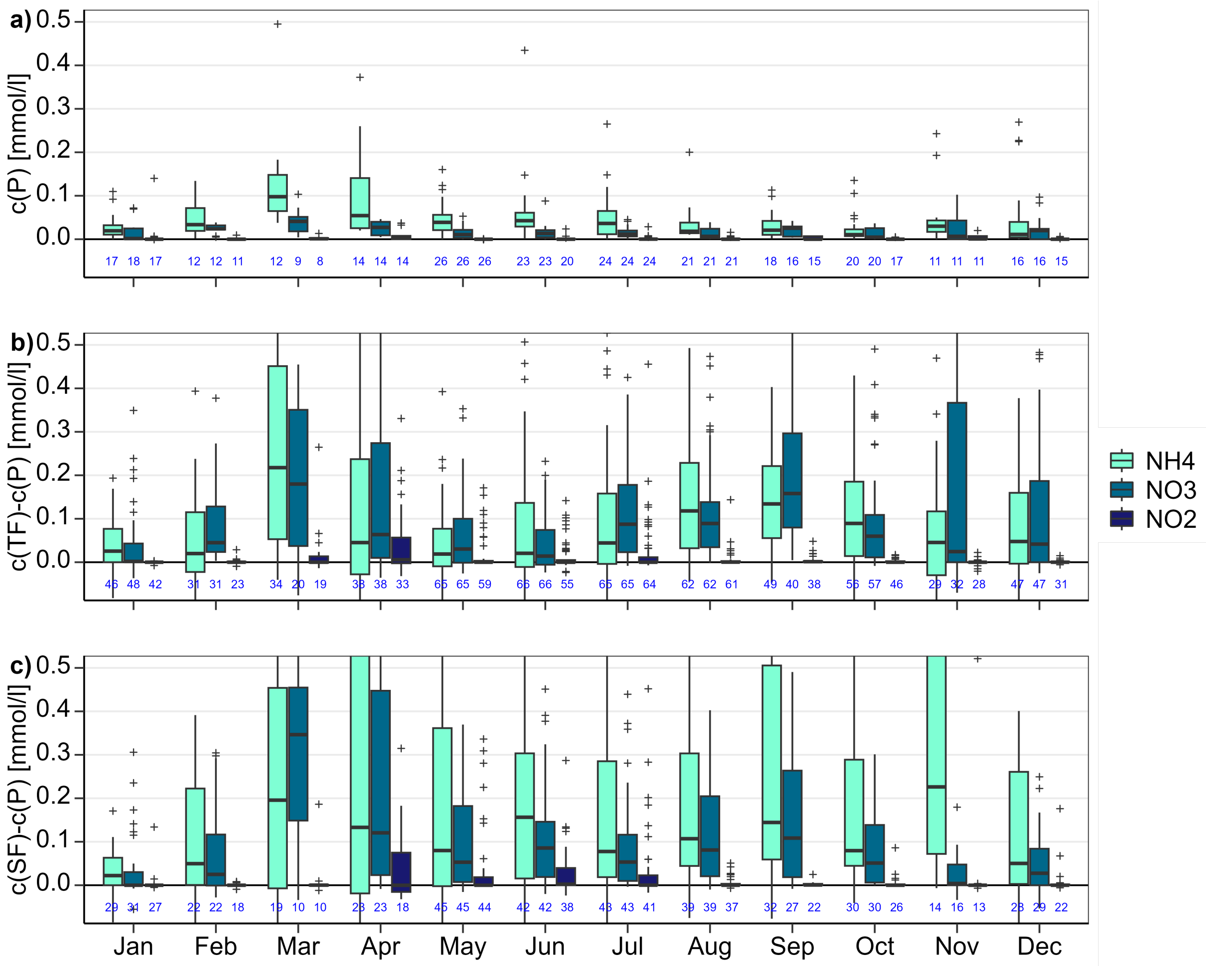


Figure 9: Seasonal variability of the concentrations of ammonium [NH_4] (light blue), nitrate [NO_3] (teal) and nitrite [NO_2] (navy) in **a)** precipitation, **b)** throughfall and **c)** stemflow.The blue numbers indicate the total number of samples used to compile the boxplots.

Nitrogen is one of the limiting factors of plant growth (Addiscott, 2005; Botter et al., 2019; Eugster and Haeni, 2013; Michel et al., 2013; Zhang, 2017). Both NH_4 and NO_3 are widely used as fertiliser, and affecting plant metabolism and biodiversity of forests they are the most important ion inputs to measure in forests ecosystems (Addiscott, 2005; Eugster and Haeni, 2013; Michel et al., 2013; Schulze, 2000). Nitrogen is thought to enter the forest mainly through dry deposition, however how much enters the forest is hard to determine since nitrogen is biologically active (Eugster and Haeni, 2013; McDowell et al., 2020). Looking at the change in concentrations of NH_4 and NO_3 after longer dry periods in figure 5b) and c) indicates that NH_4 is more influenced by dry deposition. Canopy exchange of nitrogen can go both ways: nitrogen was found both leaching from the canopy as well as taken up by the canopy (Fenn et al., 2013; D. W. Johnson and Steven E. Lindberg, 1992; Kumar Gautam et al., 2017; Rothe et al., 2002). Whether nitrogen uptake or leaching is taking place is a function of leaf or needle nitrogen content (Moffat et al., 2002). Our study site lies close to both a city and agricultural areas, and met the expectation of therefore having strong nitrogen enrichment (Michel et al., 2013). The strong enrichment in spring and autumn underline the assumption that nitrogen reaches the forest mainly through dry deposition, as these were the months where also enrichment in the dry-deposition proxy Na and Cl was highest. The high concentrations in NH_4 and NO_3 in figure 4b) and c) indicate that the high concentrations in throughfall and stemflow mainly come from the measurements taken below spruce canopies. This could either suggest that we had much higher dry deposition on the spruce as discussed in more detail in section 3.5.1 due to the rougher bark structure, or a nitrogen uptake by beech, if the nitrogen content of the leaves were lower relative to the stemflow concentration (McDowell et al., 2020). Further research is needed to determine the nitrogen fluxes and exchange along the trees and the impact thereof on the forest soil nutrient availability.

Measuring nitrogen in throughfall and stemflow increases our understanding of small scale nitrogen inputs around the stems of trees (Thimonier, Schmitt, Waldner, and Rihm, 2005), which is supposed to increase chances of soil acidification (Michel et al., 2013). Around spruce trees the large concentrations are met with often very small stemflow amounts, or for small precipitation events no stemflow at all (figure 2). The high concentrations of nitrogen could therefore origin from several precipitation events where the amount of stemflow of the previous precipitation events was not large enough to reach the forest soil. This would also explain the large variability of concentrations seen in stemflow (e.g. figure 9c)).

Interestingly we observed a rise in NO_2 in April, which we cannot explain. Nitrite usually gets produced by nitrificating bacteria in the soil, and we did not find comparable studies, as NO_2 rarely got reported in literature on throughfall and stemflow.

Overall seasonal patterns in the different ions across our site exist and it is still unclear what drives these seasonal enrichment patterns. Therefore, in the following chapter we will discuss potential forcing of enrichment patterns and their seasonality across our site.

3.5 Forcing of seasonal enrichment patterns

We found distinct seasonal differences in enrichment patterns for different ions (figure 6), however it is yet unclear to which extent these differences can be attributed to dry deposition and accumulation, canopy exchange, and dew and fog deposition, which are deemed the major enrichment sources in a forest (Lovett and S. E. Lindberg, 1984). Thus, in figure 10 we show the potential climatic forcing of enrichment seasonality across our study site. Higher precipitation intensities (figure 10a)) lead to less interception as discussed in section 3.1, which means that the concentrations measured in throughfall and stemflow will be relatively lower. Higher temperatures (figure 10b)) will increase enrichment measured in concentrations in summer due to higher evaporation, and stronger winds during spring and autumn (figure 10d)) may amplify evaporation as well as lead to more dry deposition, or input from maritime sources with low pressure systems arriving from the west during those periods. Large temperature variations around 0 °C (figure 10c)) lead to ion inputs from dew and fog along the canopies and stems. Large vapour pressure deficit such as observed during the summer months figure 10e) leads to increased transpiration by the trees and more evaporation losses. Long periods without rain (figure 10f)) might increase dry deposition, which has already been discussed in section 3.3.2.

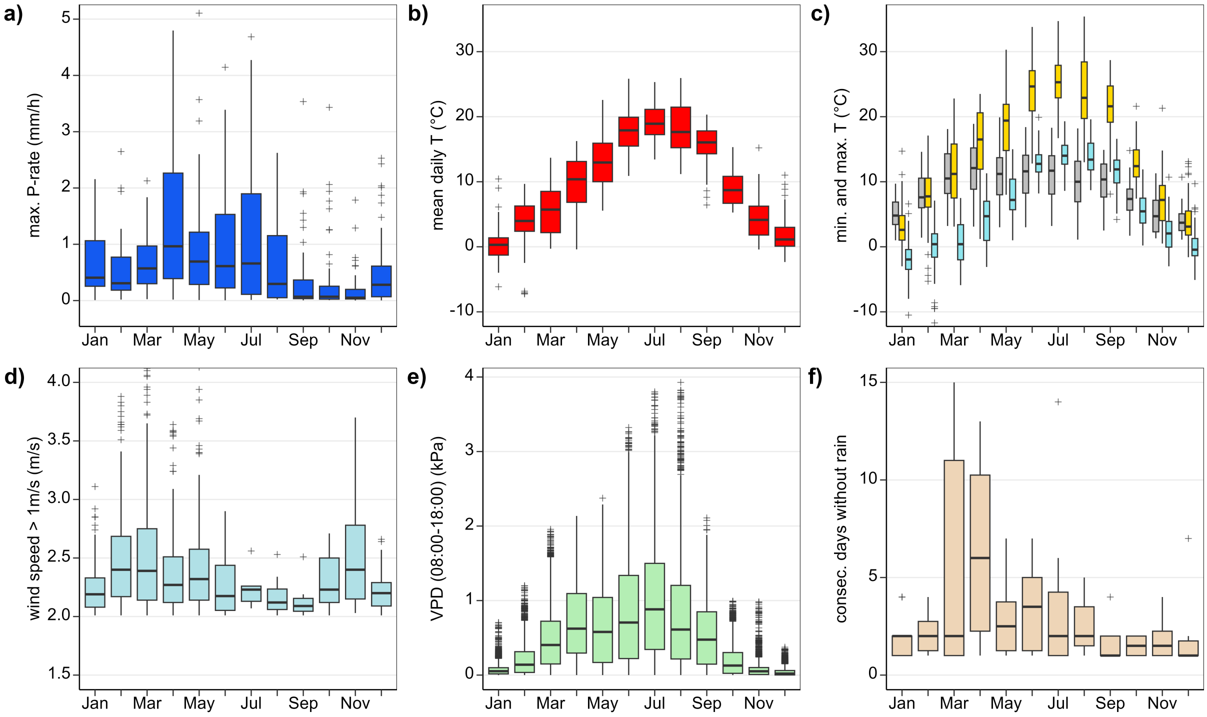


Figure 10: Potential forcing of seasonal patterns of enrichment as boxplots throughout the different months of the year. **a)** Maximum precipitation rate in mm/h, **b)** mean daily temperature in °C, **c)** minimum (blue) and maximum (yellow) daily temperature as well as the difference between maximum and minimum daily temperature (grey) in °C, **d)** mean daily wind speed if larger than 1 m/s, **e)** mean daily vapour pressure deficit (VPD) in kPa, **f)** consecutive days without rainfall.

At our site we measured ion enrichment by collecting samples after each precipitation event > 3 mm, thus signals of dry and wet deposition were mixed within the same sample, as dry deposited ions are washed out with precipitation and finally ending up in our throughfall and stemflow measuring gauges (Thimonier, Schmitt, Waldner, and Rihm, 2005). The ion concentrations in precipitation samples resemble wet deposition only, therfore the difference between the concentration in precipitation and throughfall or stemflow respectively, as shown in figures 7 to 8 can be assumed to be a measurement of dry deposition and canopy exchange.

3.5.1 Dry deposition

Dry deposition affects nutrient input into forest ecosystems by deposition of aerosols in the canopy without involvement of precipitation and fog (Andersen and Hovmand, 1999;

Lovett and S. E. Lindberg, 1984; McDowell, 1998) and is measured in our data set by the subsequent washing out of the ions from tree surfaces by precipitation. For the aerosols to settle on the tree surface, turbulence of air flow at the top of the canopy or at the forest edge are prerequisites (Adriaenssens et al., 2012; Draaijers, Van Eak, et al., 1992). Due to its high variability turbulence is hard to measure (Kumar Gautam et al., 2017), and it is further difficult to distinguish between wet and dry deposition fluxes (Staelens, Schrijver, et al., 2005) and to measure either of them separately (Macinnis-Ng et al., 2012). Thus, for dry deposition longer dry periods are beneficial (Berger, Untersteiner, et al., 2008). Due to the rougher bark structure and the accompanying larger surface area of spruce, dry deposition should be larger on spruce than on beech (Rothe et al., 2002).

During our study period, the longest periods without precipitation were recorded in in March and April, suggesting that these months are likely to experience more dry deposition enrichment. Further, the windiest periods were from February until May and in October and November, which are also periods where we observed a lot of enrichment most likely attributed to dry deposition section 3.4.1. Thus, peaks of Cl, Na, NH_4 and NO_3 in March and April and of Cl, Mg, Ca, K and NH_4 in November were potentially linked to dry deposition. However, inconsistencies exist as for example, Cl concentrations were high in November, but Na not, but they are both quoted as biologically inert. Mg, Ca and K are high in November, but not in October and December. NH_4 and NO_3 concentrations are high in March, but not so in April and also do not peak in October or November.

3.5.2 Canopy Exchange

Canopy exchange is the process of uptake or release of nutrients by trees and epiphytic vegetation over passive ion diffusion as well as gas uptake over stomata (Clark et al., 1998; Draaijers, Erisman, et al., 1997; McDowell et al., 2020; Staelens, De Schrijver, and Verheyen, 2007).

The magnitude of canopy exchange is dependent on the precipitation amount, foliage density and seasonality (Berger, Untersteiner, et al., 2008; Kumar Gautam et al., 2017; Levia and Frost, 2006). For example, nutrient values in needles of conifers are higher at the beginning and end of the growing season, suggesting that canopy leaching is small during these periods or that the leaves take up nutrients from the throughfall and stemflow (Levia and Frost, 2006; Moffat et al., 2002).

Canopy exchange is often reported for geogenic ions such as K, Mg and Ca, the direction of the exchange and relative quantities vary in literature (Adriaenssens et al., 2012; Clark et al., 1998; Macinnis-Ng et al., 2012; Parker, 1983). The nutrient content of the needles determines whether uptake or release of nutrients take place (Levia and Frost, 2006). Canopy exchange is expected to be larger on larger surface areas, therefore at beech during the growing season and at spruce all year round (Kumar Gautam et al., 2017). Also, canopy exchange via stomata is higher during high photosynthetic activity, thus when vapour pressure deficits are high (figure 10e)).

During our study period, highest solar radiation and VPD were recorded during the summer months, suggesting that these months are likely to experience more enrichment from canopy exchange, as this is also the most active vegetative season, especially for beech. Thus, the enrichment of Ca and K in May until September as well as NH_4 and NO_3 in July until September were potentially linked to canopy exchange. However, inconsistencies exist as for example, NH_4 and NO_3 concentrations were low in May and June and only increased towards the end of summer. What is still to be further researched here is the possibility of canopy uptake in the beginning of the growing season in April and May, and then possible release towards the end of it in September and October. We also found little enrichment in Ca and K in December until February, potentially indicating the smaller canopy exchange rates outside the growing season.

3.5.3 Dew and fog accumulation

High ion concentrations in dew and fog might be the reason for the peak concentrations in spring and autumn Groh, Pütz, et al. (2019), Hůnová et al. (2018), and Klemm and Wrzesinsky (2007). Being a rather overlooked water flux in mid-latitudes so far, dew might be a relevant but local and short term water input (Groh, Pütz, et al., 2019; Groh, Slawitsch, et al., 2018). Concentrations of NO_3 and NH_4 are over 10 times as large in fog compared to precipitation. Especially local effects such as local emissions may influence fog ion concentrations stronger than rain ion concentrations (Klemm and Wrzesinsky, 2007).

Dew and fog accumulation are heavily linked to relative humidity, ambient SO_2 and NO_x concentrations, air temperature and seasonality (Hůnová et al., 2018). As SO_2 concentrations were consistently small during out study, we omitted this data. However, during our study period, these periods of small VPD (figure 10e)), were recorded from October until February, low temperatures and large change in temperature (figure 10b) and c)) being recorded from March until October. This is suggesting that the months where the two overlap are likely to experience more enrichment dew and fog deposition. Thus, enrichment of NH4 and NO3 in February and March and in October and were potentially linked to dew and fog formation. However, we did not measure dew or fog throughout our study period and can therefore only make generalised assumptions, as both dew and fog are very small scale phenomena and neither their water flux contribution nor their ion contents can be determined from our data.

3.6 Conceptualisation of canopy enrichment processes

The annual enrichment pattern in the mixed temperate forest observed in this study shows distinct seasonality, which we conceptualised in figure 11. Winter (DJF) shows the lowest nutrient fluxes with no clearly distinguishable driver of the enrichment measured. Enrichment in March and April is mostly driven by dry deposition due to increased wind speeds, longer dry periods and warmer conditions. In addition to that, dew and fog deposition may lead to nutrient rich water on plant surfaces. During the summer months from May through October enrichment from dry deposition decreased while evaporative enrichment increased due to higher temperatures. Also canopy exchange processes might have played a larger role in enrichment. In November we found evidence of larger dry deposition enrichment again, combined with canopy leaching and dew and fog deposition.

The contribution of each driver separately is only a qualitative estimation. Our study highlights that seasonal variability in nutrient availability in forest ecosystems is still not well understood, thus it is of major importance to continue studies of the spatial and temporal variability of throughfall and stemflow enrichment and their forcing to assess nutrient availability in forests.

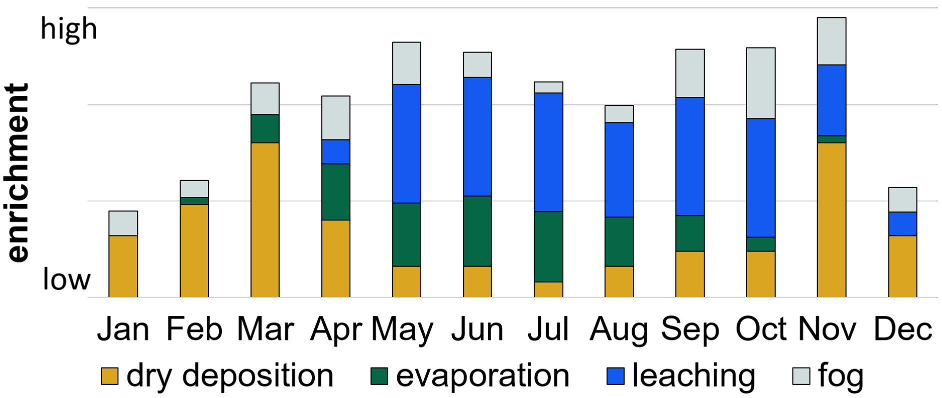


Figure 11: Conceptualisation of the annual enrichment pattern and the relative influences of the forcing dry deposition, evaporation, canopy exchange and fog inputs. We estimated the relative influence of four forcing on the annual enrichment patterns found in throughfall and stemflow in a mixed beech and spruce forest. The general annual enrichment pattern was deduced from the total annual enrichment as previously shown in figure 6. The annual evaporation pattern is based on the annual temperature as shown in figure 10b), the effect of wind was neglected. The dry deposition pattern was based on the annual pattern of Cl as shown in figure 7. The seasonality of fog was based on Hůnová et al. (2018) due to lack of sufficient data from our site. However, their study site shows a very similar climatology compared to our site.
Please note, that the effect of evaporation in the conceptual scheme might be overestimated to the loss of dry deposition.

3.7 Discussion of Uncertainties

We acknowledge that our study has obvious limitations. The findings presented above are derived from plot-scale observations within a single small forest site with only one replicate per tree species. Measurements of throughfall are challenging as there is large spatial variation, and long data series to understand temporal variability are often lacking (Brooks et al., 2012). Both of these factors may lead to large uncertainties in throughfall measurements. Longer measuring periods may mitigate the latter problem, the lack of spatial distributed information however can only be mitigated by the use of more rain gauges, an endeavour which becomes increasingly difficult if also small precipitation events are to be sampled (Levia and Frost, 2006). There are studies which mention the decrease of ion concentrations with increasing distance from the stem (Adriaenssens et al., 2012), a property of throughfall which makes studies performed with single rain gauges highly sensitive to the placement of the rain gauge. Thus, in our study, rather than using multiple gauges across the site, we focused on integrated measurement of throughfall (with gutters) directly below the canopies of three stands, however we could collect a reasonably high resolved and long time series i.e., each precipitation event > 3 mm for almost three years. For inter study comparison, reporting of nutrient fluxes in either mg/l and mmol/l is common. While the former is easier for mass flux calculations, we chose the latter for this study to be able to conduct stochastic comparisons such as performed in section 3.4.1, which makes it hard to determine the absolute nutrient influx from throughfall or stemflow. Thus, although many of our results are suggestive rather than definitive, they point to the need for further research on the water and nutrient inputs below different tree species canopies to better understand water and nutrient dynamics in forest soils.

4 Conclusions

The water and nutrient availability for a forest ecosystem mainly depend on the tree species composition and precipitation intensity which determine the amount and concentration of throughfall and stemflow. Interception patterns are different across seasons and for tree species (i.e., spruce and beech) and not purely related to canopy cover but also seasonal differences in precipitation intensities. While interception under coniferous, evergreen tree species is mainly affected by precipitation patterns, interception below deciduous tree species is also affected by the seasonality of canopy cover. In percentage of total annual precipitation, stemflow is negligible, however it might be a relevant water input on small scales i.e., around the stem and to the root system.

Likewise, ion enrichment in throughfall and stemflow has seasonal variation, that can not solely be explained by dry deposition. Although concentrations were mostly higher after drier antecedent periods, dry deposition is not the only driver of ion enrichment in throughfall and stemflow. However, evaporative enrichment, canopy exchange, and dew and fog deposition yield major effects on the seasonality of iron enrichment. While dry deposition is increased by longer dry periods and stronger winds and is therefore prevalent in spring and autumn, evaporative enrichment is important during the summer months. Canopy leaching is an important driver during the growing season, however it is dependent on tree type and on the seasonal activity of nutrient uptake of the tree. Dew and fog deposition may lead to peaks in ion enrichment in spring and autumn. Also, enrichment in throughfall and stemflow is not statistically different for most ions.

Overall, our study highlights the complex interactions between tree species and climate forcing that affect the seasonal variability in water and nutrient supply to forest ecosystems.

Acknowledgement

We thank the team of the research assistants at the Waldlabor Zurich: Lena Straumann, Lucas Truniger, Raphael Meier, Anna Lena Könz, Lara Virsik, Fabian Strittmatter, Elisa Hage, Marc Bühler and Gian-Luca Cavelti for their fantastic work in collecting precipitation samples, repairing stemflow raingutters and reading out sensors. We further thank Stefano Martinetti from the research group supporting us in the field and Lucien Biolley supporting us in the laboratory.

A Supplement

A.1 Canopy Cover - Threshold function

```
"autoThreshold" <- ### function from the former package tiff which
    is discontinued for R4.0.0 or later, therefore implemented
   here
  function(d.m, est=0.5) {
    est.old <- 0
    while (est.old != est) {
      est.old <- est
      t1 <- mean(d.m[d.m < est], na.rm=TRUE)
      t2 <- mean(d.m[d.m > est], na.rm=TRUE)
      est <- mean(c(t1, t2), na.rm=TRUE)
    }
    return(c(t1, mean(c(t1, est)), est, mean(c(t2,est)), t2))
  }
```

A.2 Equations

$$IC_{all} = 77.7 - 4.6 \cdot \log(\mathrm{PR}) \tag{2}$$
$$IC_B = 67.6 - 4.6 \cdot \log(\mathrm{PR}) \tag{3}$$
$$IC_{mS} = 78.8 - 4.8 \cdot \log(\mathrm{PR}) \tag{4}$$
$$IC_{yS} = 85.0 - 3.9 \cdot \log(\mathrm{PR}) \tag{5}$$

$$DC_{B,\ Hemery} = 15.23 \cdot DBH + 1.13312 \tag{6}$$
$$DC_{B,\ Sharma} = 15.0 \cdot DBH + 1.4 \tag{7}$$
$$DC_{S,\ Sharma} = 8.7 \cdot DBH + 1.4 \tag{8}$$

A.3 Tables

Table 1: P-values of paired Wilcox tests comparing the difference in the amounts of throughfall and stemflow. The difference between all 5 datasets are significant with p-values < 0.05.

	TF1	TF2	TF3	SF1	SF2
TF1	1	2.70E-03	5.94E-08	2.03E-25	1.54E-25
TF2	-	1	5.75E-03	8.81E-21	6.00E-24
TF3	-	-	1	3.05E-12	2.00E-13
SF1	-	-	-	1	6.73E-15
SF2	-	-	-	-	1

Table 2: Significance of trend analysis for a decreasing trend in concentration values for higher antecedent precipitation, performed with a Mann-Kendall-test on data from precipitation, throughfall and stemflow water samples sorted by the amount of 10 day antecedent precipitation.

water samples	**name**	**p.value**	**significance**
precipitation	Ca	1.24 E-01	-
	Cl	2.08 E-03	< 0.05
	K	2.78 E-02	< 0.05
	Mg	7.15 E-04	< 0.05
	Na	3.48 E-01	-
	NH4	2.80 E-02	< 0.05
	NO2	1.28 E-01	-
	NO3	5.21 E-02	-
throughfall	Ca	1.85 E-16	< 0.05
	Cl	5.46 E-21	< 0.05
	K	1.71 E-13	< 0.05
	Mg	2.89 E-19	< 0.05
	Na	3.81 E-06	< 0.05
	NH4	2.07 E-13	< 0.05
	NO2	1.13 E-01	-
	NO3	1.15 E-05	< 0.05
stemflow	Ca	1.76 E-03	< 0.05
	Cl	6.39 E-12	< 0.05
	K	1.40 E-04	< 0.05
	Mg	1.40 E-07	< 0.05
	Na	5.28 E-02	-
	NH4	1.78 E-05	< 0.05
	NO2	1.41 E-01	-
	NO3	2.20 E-02	< 0.05

Table 3: Significance of differences between concentrations in throughfall and stemflow for the ions calculated with a paired Wilcox test.

datasets	name	p-value	significance
TF/SF	Ca	5.6E-05	< 0.05
TF/SF	Cl	9.5E-01	-
TF/SF	K	8.9E-05	< 0.05
TF/SF	Mg	9.5E-01	-
TF/SF	Na	2.2E-01	-
TF/SF	NH4	3.6E-05	< 0.05
TF/SF	NO2	1.9E-01	-
TF/SF	NO3	6.5E-01	-

Table 4: Significance of differences between ion concentrations in stemflow and throughfall when comparing the concentration measured under different trees (beech B, mature spruce mS and young spruce yS).

water samples	comparison	ions	p-value	significance
SF	B/mS	Ca	9.3E-11	< 0.05
	B/mS	Cl	7.8E-10	< 0.05
	B/mS	K	1.7E-22	< 0.05
	B/mS	Mg	1.9E-06	< 0.05
	B/mS	Na	4.1E-19	< 0.05
	B/mS	NH4	1.0E-17	< 0.05
	B/mS	NO2	1.7E-01	-
	B/mS	NO3	1.0E-06	< 0.05
TF	B/mS	Ca	6.0E-02	-
	B/mS	Cl	1.9E-08	< 0.05
	B/mS	K	5.3E-09	< 0.05
	B/mS	Mg	3.0E-02	< 0.05
	B/mS	Na	2.9E-06	< 0.05
	B/mS	NH4	3.2E-10	< 0.05
	B/mS	NO2	1.9E-01	-
	B/mS	NO3	2.5E-07	< 0.05
TF	B/yS	Ca	8.0E-01	-
	B/yS	Cl	2.9E-08	< 0.05
	B/yS	K	8.2E-12	< 0.05
	B/yS	Mg	5.5E-03	< 0.05
	B/yS	Na	1.1E-02	< 0.05
	B/yS	NH4	1.1E-07	< 0.05
	B/yS	NO2	4.7E-01	-
	B/yS	NO3	2.6E-07	< 0.05
TF	mS/yS	Ca	1.1E-01	-
	mS/yS	Cl	8.6E-01	-
	mS/yS	K	3.1E-01	-
	mS/yS	Mg	6.0E-01	-
	mS/yS	Na	2.5E-02	< 0.05
	mS/yS	NH4	6.3E-01	-
	mS/yS	NO2	5.5E-01	-
	mS/yS	NO3	5.7E-01	-

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