Atmospheric deposition of elements and its relevance for nutrient budgets of tropical forests

Title

Atmospheric deposition of elements and its relevance for nutrient budgets of tropical forests

Authors

Leandro Van Langenhove[a], Lore Verryckt[a], Laëtitia Bréchet[a], Elodie A. Courtois[b], Clement Stahl[c], Florian Hofhansl[d], Marijn Bauters[e], Jordi Sardans[f,g], Pascal Boeckx[e], Erik Fransen[h,i], Josep Peñuelas[f,g], Ivan A. Janssens[a]

[a] Centre of Excellence PLECO (Plants and Ecosystems), Department of Biology, University of Antwerp, Wilrijk, Belgium
[b] Laboratoire Ecologie, évolution, interactions des systèmes amazoniens (LEEISA), Université de Guyane, CNRS, IFREMER, French Guiana, 97300 Cayenne, France
[c] INRA, UMR EcoFoG, CNRS, Cirad, AgroParisTech, Université des Antilles, Université de Guyane, 97387 Kourou, France
[d] International Institute for Applied Systems Analysis (IIASA); Schlossplatz 1, A-2361 Laxenburg, Austria.
[e] Isotope Bioscience Laboratory–ISOFYS, Ghent University, 9000 Gent, Belgium
[f] CSIC, Global Ecology Unit CREAF-CSIC-UAB,08193 Bellaterra, Catalonia, Spain
[g] CREAF, 08193 Cerdanyola del Vallès, Catalonia, Spain
[h] Center of Medical Genetics, University of Antwerp, Prins Boudewijnlaan 43/6, Edegem, BE-2650 Antwerp, Belgium
[i] StatUa Center for Statistics, University of Antwerp, Prinsstraat 13, BE-2000 Antwerp, Belgium

Abstract

Atmospheric deposition is an important component of the nutrient cycles of terrestrial ecosystems, but field measurements are especially scarce in tropical regions. In this study we analysed 15 months of precipitation chemistry collected in an old growth tropical forest located in French Guiana. We measured nutrient inputs via bulk precipitation and throughfall and used the canopy budget model to estimate nutrient fluxes via canopy exchange and dry deposition. Based on this method we quantified net fluxes of macronutrients and compared their contribution to internal cycling rates via litterfall. Our results suggest that while atmospheric deposition of nitrogen was relatively high (13 kg ha^{-1} y^{-1}), and mainly in organic forms, the N inputs via litterfall were an order of magnitude higher. In contrast to nitrogen, we found that atmospheric deposition of phosphorus (0.5 kg ha^{-1} y^{-1}) supplied up to one third of the annual litterfall input to the forest floor. Most strikingly, combined annual inputs of potassium via atmospheric deposition (14 kg ha^{-1} y^{-1}) and canopy leaching (22 kg ha^{-1} y^{-1}) were three times larger than internal nutrient recycling via litterfall (11 kg ha^{-1} y^{-1}). We conclude that atmospheric deposition of phosphorus and especially potassium may play an important role in sustaining the productivity of this old-growth tropical rainforest.

Keywords

Throughfall, Litterfall, Nutrient Cycling, Nitrogen, Phosphorus, Potassium

Introduction

Tropical forests are of major significance to the global carbon budget as they account for 70% of the global gross forest carbon sink and about 55% of the global forest carbon pool is stored in tropical forests (Pan et al. 2011). In order to sustain the high productivity and carbon sink-strength of tropical forests, nutrient inputs are needed to compensate for the continuous nutrient losses by biomass removal, leaching, erosion, or denitrification (Vitousek 1984; Wieder et al. 2015). Three main natural pathways add nutrients to ecosystems, namely mineral weathering, Nitrogen (N) fixation (only for N) and atmospheric deposition. Over short timescales, weathering rates are thought to be slow in lowland tropical forests (Porder et al. 2005) as they are typically characterized by already highly weathered soils and little altitudinal variation, which leads to slow rates of soil rejuvenation through erosion (Baillie 1996; Vitousek et al. 2010; Walker and Syers 1976). In mature lowland tropical forests the input of external N through free-living (soil, leaf litter) or symbiotic N fixation is highly variable and both high and low rates have been reported (Reed et al. 2007; Reed et al. 2011; Sullivan et al. 2014; Taylor et al. 2019; Van Langenhove et al. 2019; Wurzburger et al. 2012). Possibly, atmospheric deposition represents an important external nutrient input for these tropical forests where other external inputs, such as rock weathering (Houlton et al. 2018) or N fixation, are low (Pacyna 2008) and could therefore be a key component of their nutrient budget (Hofhansl et al. 2012). Unfortunately, there is a particular gap in atmospheric deposition data from tropical forests (Jia et al. 2016; Vet et al. 2014) and the projected rise in atmospheric deposition of nutrients, such as N and phosphorus (P) mainly due to anthropogenic activities, in tropical regions highlights the need for empirical studies on this topic (Galloway et al. 2004; Wang et al. 2017).

Since the 1960s, the most common method to quantify atmospheric deposition to ecosystems is capturing and chemically analysing bulk precipitation, revealing the composition of the precipitation (Hofhansl et al. 2011; Whitehead and Feth 1964). Various sources contribute to atmospheric gases and particles, including natural (e.g. sea spray and soil dust) and anthropogenic (e.g. fossil fuel combustion, biomass burning, agriculture and fertilizer production) emission sources (Deusdará et al. 2016). Gases and aerosols form cloud condensation nuclei or are scavenged by raindrops (Hobbs 2000). It is important to identify the chemical composition of bulk precipitation and the sources of elements in rainwater to understand the regional dispersion of nutrient input and its potential impacts on ecosystem functioning (Mphepya et al. 2004; Xiao et al. 2013).

Besides atmospheric nutrient inputs into the ecosystem, nutrients are recycled via production and decomposition of dead plant material and through dissolution of elements in throughfall (TF) and stemflow. The contribution of root turnover and decay to internal nutrient cycling remains poorly understood in terrestrial ecosystems in general (See et al. 2019), but the annual input of aboveground litterfall has been well characterised in a variety of tropical forests (Chave et al. 2010) as it represents a major pathway for nutrient input (e.g. N, P and sulphur (S)) to the forest floor. However, litterfall does not represent an external nutrient input to the ecosystem, but rather represents an internal recycling of nutrients, in contrast to nutrients released through weathering or brought in via atmospheric deposition. Throughfall, defined as the precipitation that falls through a forest canopy or comes in contact with the canopy and falls to the forest floor (Bales et al. 2011), represents a combination of atmospheric deposition of nutrients and internal recycling processes. Three pathways combine to form throughfall (Ulrich 1983): (I) passage of incident precipitation (bulk precipitation) through the canopy, (II) wash-off of dry deposited materials (dry deposition),

such as particles and adsorbed gases, and (III) bi-directional solute exchange between intercepted rainfall and canopy surfaces like foliage, woody parts, epiphytes and microorganisms (canopy exchange). Net throughfall is thus affected by bulk precipitation, dry deposition and canopy exchange. Separating the contribution of dry deposition from canopy exchange in throughfall has been an ongoing concern for several decades (Staelens et al. 2008), but is commonly calculated via the canopy budget model (Bredemeier 1988; Draaijers et al. 1996; Ulrich 1983). This model distinguishes between internal and external input sources to ecosystems using the so-called 'filtering approach' based on a tracer ion (Ulrich 1983) and by estimating ion exchange processes occurring within the forest canopy (Draaijers et al. 1996).

Although the past two decades have seen an increase in research linking patterns of rainfall and plant productivity in lowland tropical forests (Taylor et al. 2017) there is still a lack of studies on atmospheric deposition in areas of high precipitation and temperature (Jia et al. 2016). Throughfall measurements from South American tropical forests are especially rare (Bauters et al. 2019) and Vet et al. (2014) revealed that long-term measurements are even rarer, with only a handful of studies reporting rainfall chemistry for periods of one year or longer in the neotropics (excluding Central America).

Here, we present the results of a study spanning 15 months and report on annual fluxes of bulk deposition and throughfall in a tropical forest in French Guiana. We compared the input of external nutrients by bulk deposition to the internal recycling by litterfall and throughfall. For elements such as phosphorus (P) and potassium (K), that have often been reported to be limiting in tropical rain forests (Sardans and Peñuelas 2015; Turner et al. 2018; Wright et al. 2011), we expected a relatively large amount of bulk deposition compared to the litterfall and

throughfall P and K. Specifically, we aimed to (I) quantify the annual deposition of elements by bulk precipitation, (II) assess to what degree the canopy altered these fluxes, and (III) compare the external nutrient input to the nutrients cycling via litter fall.

Materials and Methods

Site description

The study was conducted at the Paracou research station (5°15'N, 52°55'W), 15 km from the coast and about 40 km West from the nearest city, Kourou, in French Guiana. Annual rainfall (2004 - 2015) averaged 3100 ± 70 mm yr^{-1} and annual air temperature 25.7 ± 0.1 °C (Aguilos et al. 2019). The tropical wet climate is highly seasonal due to the north/south movement of the Inter-Tropical Convergence Zone, which brings heavy rains from December to July and a very dry period from August to November (less than 100 mm precipitation per month). The vegetation is highly diverse, with between 150 and 200 species ha^{-1} for trees with diameter at breast height > 10 cm. The most represented plant families occurring at the study site are the Lecythidaceae, Fabaceae, Sapotaceae and Chrysobalanaceae (Gourlet-Fleury et al. 2004). Soils in Paracou are mostly acrisols (FAO 1998) developed over a Precambrian metamorphic formation called the Bonidoro series. It is characterized by schists and sandstones and locally crossed by veins of pegmatite, aplite and quartz (Epron et al. 2006). The landscape is undulating with maximum slopes of approximately 15°. The elevation difference between hill summits and valleys is 20 - 50 m over horizontal distances of 200 - 400 m.

Experimental setup

Bulk and Throughfall precipitation

Precipitation collectors were made of polyvinyl chloride (PVC) funnels (202 mm diameter, area = 320.47 cm^2) attached to an opaque PVC plastic tube (diameter 15 mm) inserted into a 5 L polyethylene (PE) plastic bottle. Bottles were covered with aluminium foil, inserted into a 0.5

m deep pit and, for the bulk precipitation collectors only, a wooden lid covered with aluminium foil was installed over each pit to limit solar heating. A small hole was drilled into the lid to allow the plastic tube passing through. For the bulk precipitation, three blocks of eight collectors each were set up in a forest clearing nearby the Paracou research station. Between-block distances were approximately 20 m. For the throughfall, three blocks of eight collectors each were set up underneath the forest canopy within a 300 m radius of the Paracou flux tower (Bonal et al. 2008). For both bulk precipitation and throughfall, the eight collectors within a block were spaced 1.5 - 2 m apart in two rows of four collectors.

Collectors were installed mid-august 2016 and, starting on September 2nd 2016, sampling was carried out every two weeks and both bulk precipitation and throughfall collectors were sampled on the same day. To avoid collectors overflowing during periods of heavy rains (December 2016 - February 2017 and April 2017 - June 2017), collectors were sampled more frequently and the time between sampling was reduced to one week. Because of extremely heavy rainfall in May 2017, we reduced the time between sampling to three days for two sampling events. At each sampling event, the total precipitation collected in each bulk precipitation and throughfall collector bottle was measured and recorded in the field with a graduated cylinder (accuracy 5 mL when volume in bottle exceeded 1 L and 1 mL accuracy when volume in bottle was below 1 L). Collected water was pooled together per block and an aliquot of 150 mL was subsampled. This sample was brought to the lab on the same day the sampling was carried out and immediately frozen at -20 °C.

Littertraps

Litter collectors were made of 40 x 70 cm (area = 0,28 m²) polypropylene (PP) plastic nets strung up between 1.2 and 1.5 m above the forest floor and attached to four nearby trees with strings (each net corner attached to one tree). We used a bubble leveller to make sure

each net surface was level. We installed twelve blocks of nine nets per block (N = 108) in the vicinity of the Paracou flux tower, in the same locations as described in Courtois et al., (2018). Traps within a block were spaced 15 m apart in a circular pattern with one littertrap in the centre. Nets were emptied every three weeks, with the exception of August and September when they were emptied every two weeks because of expected higher litterfall at the onset of the dry season (Chave et al. 2010). Each time the nets were emptied the contents of each littertrap, with the exception of larger branches (length > 30 cm and / or diameter > 2 cm) that were discarded, were gathered in a paper bag and brought to the lab where bags were first dried at 70 °C (48 h) and then weighed. Afterwards, litter from a single block was pooled together, mixed by hand during two minutes and subsampled (approximately 20 g).

Chemical analyses

Chemical analyses were performed at the Ecosystem Management Research Group, University of Antwerp, Belgium. Samples of the bulk precipitation and throughfall were defrosted and an aliquot was filtered (0.20 µm) preceding the analysis of inorganic anions (Cl^-, NO_3^-, SO_4^{2-}, HPO_4^{2-}) and cations (Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}) by ion chromatography on an 883 Basic IC Plus (MetrOhm, Switzerland) with 919 IC Autosampler Plus (MetrOhm, Switzerland). Nitrite (NO_2^-) was also measured in these samples, but concentrations were consistently below 1 µg L^{-1} and are not discussed further. Anions were separated on a Metrosep A Supp 5 - 150 / 4.0 column with a Metrosep A Supp 5 guard / 4.0 protecting the separation column. Cations were separated on a Metrosep C 6 - 150 / 4.0 column with a Metrosep C 4 guard / 4.0 protecting the separation column. All columns were produced and distributed by MetrOhm, Switzerland. Data analysis was conducted with MagicIC Net software. For the determination of total dissolved nitrogen (TDN) and total dissolved phosphorus (TDP) we used the Kjeldahl

159 method where samples were destructed using a mixture of potassium sulfate, sulfuric acid
160 and selenium, heated to 400 °C for 90 minutes (ISO, 1984). This converts all N and P containing
161 compounds into NH_4^+ and HPO_4^{2-}, which are subsequently measured on a SAN++ Continuous
162 Flow Analyser (Skalar, The Netherlands). Following this, TDN is calculated by adding the
163 measured NH_4^+ from the SAN++ CFA to the NO_3^- measured by the ion chromatography, DON
164 is calculated by subtracting the NH_4^+ measured by the ion chromatography from the NH_4^+
165 measured by the SAN++ CFA and DOP is calculated by subtracting the HPO_4^{2-} measured by ion
166 chromatography from HPO_4^{2-} measured by the SAN++ CFA.

167 Dried litter samples were homogenized in a ball mill (Retsch MM2000) and aliquots (1.5 – 2
168 mg) were weighed into tin capsules for analysis of total carbon and nitrogen content by
169 elemental analysis (Flash 2000, Thermo Fisher Scientific, Germany). For phosphorus and
170 cation analysis, litter samples were first acid digested (Walinga et al. 1995). Approximately
171 300 mg dried samples were wet digested with 2.5 mL of a salicylic acid and sulphuric acid-
172 selenium mixture in 25 mL glass flasks on a heating plate at 100 °C. After 2 h samples were
173 allowed to cool down and three times hydrogen peroxide (1 mL) was added. Next, the
174 temperature was increased to 330 °C and samples were left to stand until digests had turned
175 colourless after which they were cooled down, mixed in 48.3 mL of high purity water and
176 allowed to settle overnight. Cations (K^+, Ca^{2+}, Mg^{2+}) were measured by Inductively Coupled
177 Plasma Optical Emission Spectroscopy using the iCAP6300 Duo ICP-OES (Thermo Fischer
178 Scientific, Germany) and total phosphorus was measured on a SAN++ Continuous Flow
179 Analyser (Skalar, The Netherlands).

Definitions and calculations

The volume weighted mean (VWM, Eq. 1) concentration (mg L^{-1}) of each ionic rainwater component was used to express solute concentration of bulk precipitation (BP) and throughfall (TF) during the study period and was calculated as follows:

$$\mathrm{VWM} = \frac{\sum(\mathrm{C_i} * \mathrm{V_i})}{\sum(\mathrm{V_i})} \quad (1)$$

Here, C is the ionic concentration in mg L^{-1} measured for each block *i* per sampling event and V is the measured rainfall volume in mm of each block *i* for each sampling event.

Next, we multiplied the measured concentration of each ion (C_i) within a block with the sampled rainfall volume (V_i) from that same block and rescaled the unit to kg ha^{-1} (Eq. 2). We then obtained the annual nutrient flux (ANF) in kg ha^{-1} y^{-1} by summing the means of all three blocks per sampling event (j) within one year (September 2016 - September 2017). We did this for all ions from both bulk precipitation and throughfall.

$$\mathrm{ANF} = \sum_{\mathrm{j}}^{\mathrm{n}} \mathrm{mean}(\mathrm{C_i} * \mathrm{V_i})_{\mathrm{j}} \quad (2)$$

Throughfall is the amount of bulk precipitation that is not retained by the vegetation but passes through the canopy layer and is thus altered in hydrochemistry due to canopy exchange (CE) and dry deposition (DD) (Eq. 3):

$$TF = BP + CE + \mathrm{DD} \quad (3)$$

Net throughfall flux is defined as the difference between the solute flux in throughfall and in bulk precipitation (Eq. 4):

$$\mathrm{NTF} = \mathrm{TF} - \mathrm{BP} = \mathrm{CE} + \mathrm{DD} \quad (4)$$

To estimate the contribution of canopy exchange and dry deposition to annual net throughfall fluxes we followed the canopy budget model (Ulrich 1983).

Canopy Budget Model

The canopy budget model simulates the interaction of substances within forest canopies based on measurements of throughfall and precipitation and is therefore applicable to distinguish the relative importance of dry deposition and canopy exchange to net throughfall fluxes. The model has been used to estimate dry deposition and canopy exchange fluxes in a wide range of forest ecosystems, and different approaches have been reported (Hofhansl et al. 2011; Staelens et al. 2008). Here we used the filtering approach of the canopy budget model (Ulrich 1983) which is commonly used to estimate dry deposition. In this approach aerosols are assumed to be deposited with equal efficiency onto the forest canopy as particles containing an inert tracer ion. Sodium is often used as a tracer ion because it is assumed not to be influenced by canopy exchange processes (Staelens et al. 2008), however, both modest canopy leaching and uptake of Na^+ has been reported (Tobón et al. 2004; Wanek et al. 2007). The calculations are commonly based on mean (semi)annual fluxes in bulk deposition and throughfall per forest site. Dry deposition of a certain element X (DD_X) is calculated as the product of the dry deposition factor of the tracer ion (here: Na^+), which is the NTF to BP ratio of Na^+, times the BP of element X (Eq. 5)

$$DD_X = \frac{(TF_{Na} - BP_{Na})}{BP_{Na}} * BP_X \quad (5)$$

Canopy exchange is then determined by subtracting dry deposition from NTF (Eq. 4).

Sea salt fraction

A common method used to quantify the inputs of marine chloride (Cl^-), sulfate (SO_4^{2-}) magnesium (Mg^{2+}), calcium (Ca^{2+}) and potassium (K^+) in rainwater is to compare the ionic

ratios of the rainwater with the ionic ratios of the seawater using sodium (Na^+) as the reference ion. The chemical concentrations exceeding the ionic ratio of seawater, known as non-sea-salt (nss) fraction, were estimated as follows by assuming that all Na^+ is from marine origin (Berner and Berner 2012):

$$[\mathrm{X}]_{\mathrm{nss}} = [\mathrm{X}]_{\mathrm{rain}} - \left([\mathrm{Na^+}]_{\mathrm{rain}} \times \left[\frac{\mathrm{X}}{\mathrm{Na^+}}\right]_{\mathrm{SW}}\right) \quad (8)$$

Where $[X]_{nss}$ is the non-sea-salt fraction of the ion X, $[X]_{rain}$ and $[Na^+]_{rain}$ are the measured concentrations of the ion X and Na^+ in the rainwater, respectively, and $\left[\frac{X}{Na^+}\right]_{SW}$ is the ratio of the concentrations of X to Na^+ in the seawater (all components are expressed in mg L^{-1}). The nss fractions were used to calculate the ionic concentrations in the rainwater that resulted from non-marine inputs.

Statistical analysis

We calculated the coefficient of variation (CV) of the nutrient concentrations between the three distinct bulk precipitation and throughfall setups for each sampling event to look the variation between the funnel setups. We used Pearson correlations to look at the correlations between bulk precipitation and throughfall volume and each nutrient concentration and flux. Differences in ion fluxes between atmospheric deposition (the sum of bulk precipitation and dry deposition) and throughfall, i.e the canopy exchange, were investigated using linear mixed effects regression (LMER) models. Because in all cases the data were right skewed they were transformed using a Box-Cox analysis to find the optimal transformation. After transformation, normality was confirmed visually by inspecting the histograms and qq-plots. The data of four of the 13 measured ions was zero inflated and the occurrence of zeroes ranged between 12.5 and 58%. In these cases, we split the analysis into two parts. In the first part we constructed a new binomial vector containing information on the presence or absence

of the ion of interest. We then used a generalized linear mixed effects (GLMER) model with setup (atmospheric deposition or throughfall) as fixed effect and sampling date as random effect to test if the occurrence of zeroes differed between the bulk precipitation and throughfall. For the second part we removed the zeroes from the dataset and applied LMER models to see if, in the cases there is measurable ion flux, there were differences between bulk precipitation and throughfall. Again we entered setup (bulk precipitation or throughfall) as fixed effect and sampling date as random effect. Analyses were carried out in R (R Core Team 2018) with the package lmerTest (Kuznetsova et al. 2017). For visualisations we used the ggplot2 package (Wickham 2016).

Results

Bulk precipitation

Between August 19th 2016 and November 15th 2017 we measured a bulk precipitation of 3798 ± 88 mm in the forest clearing collectors. For one year, between September 23rd 2016 and September 20th 2017, the bulk precipitation was 3550 ± 86 mm. A peak in monthly precipitation was measured in May 2016, with 765 ± 12 mm rainfall. In both 2016 and 2017, bulk precipitation in August, September, October and November remained below 100 mm month^{-1} (Fig 1). These months are typical dry season months.

The spatial coefficient of variation (CV) of bulk precipitation, calculated for the three blocks and for each sampling period separately, ranged between 0.5 and 53.3%. Mean CV was 9.7% and the median was 3.9%. There was no relationship between the precipitation volume and the deviation from the mean.

There was variation in bulk precipitation ion concentration between the three collector setups, with average CVs (N = 3) ranging between 14 and 40% for most ions, except for NH_4^+

and both inorganic and organic P, which had higher CVs. (Table 1). Nutrients that overall had very low concentrations, such as dissolved organic P (DOP), also exhibited the highest CVs, likely because their measured concentrations were often just above or below the quantification limit.

The ion concentrations of all measured ions correlated negatively with the amount of precipitation, albeit with large variation in correlation strength between the nutrients. Conversely, with the exception of DOP, the ion flux (kg ha^{-1} y^{-1}) of all measured ions correlated positively with the amount of precipitation (Table 2).

There were large differences in concentrations and fluxes among the different ions over the study period (Table 3). Given that concentrations and fluxes are mathematically inter-related, we will only present and discuss the ion fluxes here. Bulk precipitation flux of TDP was low, amounting to 0.4 kg ha^{-1} y^{-1}, while TDN deposition was much higher, amounting to 10.5 kg ha^{-1} y^{-1}, and resulting in a molar N:P input ratio of 51. Inorganic phosphorus (P_i) accounted for most (79%) of the deposited P, while N_i, here defined as the sum of NO_3^- and NH_4^+, accounted for only 17% of the deposited N. Fluxes of SO_4^{2-}, Na^+ and Ca^{2+} were relatively high, all exceeding 25 kg ha^{-1} y^{-1}, while the deposition flux of Cl^- was extremely high at 120 kg ha^{-1} y^{-1} (Table 3).

The percentage of each ion that was derived from sea salt (ss fraction) showed large variation (Fig 2). Because we assumed it to be completely derived from sea salt (Berner and Berner 2012), the ss fraction of Na^+ was 100%. Surprisingly, the ss fraction of Cl^- was less than 50%, indicating a large non-marine input. The ss fraction of Mg^{2+} exceeded 80%, while the ss fractions of SO_4^{2-}, K^+ and Ca^{2+} were below 25%. We did not calculate the ss fraction of the N

and P forms because they are not derived from sea salt and thus generally assumed to be completely derived from other sources (Berner and Berner 2012).

Throughfall and canopy exchange

Over the study period throughfall was 2834 +- 76 mm (Fig 1). Compared to bulk precipitation, rainfall interception by the forest canopy was thus 25%. Throughfall volumes correlated well with bulk precipitation volumes (Pearson r = 0.982, Fig 3a) and higher bulk precipitation volumes correlated with increased canopy interception (Pearson r = 0.814, Fig 3b). Similarly to bulk precipitation, we found that the concentration of most ions (Cl^-, NO_3^-, P_{in}, SO_4^{2-}, NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+}, TDN and DON) in the throughfall correlated negatively with throughfall volume and, vice versa, fluxes correlated positively to throughfall volume for most nutrients (with the exception of NH_4^+, TDP and DOP, Table 2).

Generally, variations in nutrient concentration among the three setups underneath the canopy were high, with CVs ranging between 12 and 80%. Overall, the ion concentrations in throughfall were slightly higher than the concentrations in bulk precipitation, with the exception of NO_3^- and P_{in} (Table 3). However, given that overall throughfall volume was 25% lower than bulk precipitation, the throughfall flux of Cl^-, NO_3^-, P_{in}, SO_4^{2-} and Ca^{2+} to the forest floor was smaller than the amount deposited by bulk precipitation. Following the canopy budget model, the dry deposition, which was calculated based on the Na^+ enrichment after passage through the canopy (eq. 5), averaged at 26% of the wet precipitation across all ions.

The canopy exchange of Cl^-, SO_4^{2-}, Ca^{2+}, NO_3^-, P_{in} and TDP was negative, implying that there was net uptake of these nutrients by the canopy (Table 3, Fig 5). Potassium, in contrast, behaved very differently and was leached from the canopy at a rate of 22 kg ha^{-1} y^{-1} (Fig 5). No net canopy exchange was observed for Mg^{2+}, NH_4^+, DON, TDN and DOP, indicating that

either there was no canopy uptake or leaching of these nutrients, or that these processes balanced each other out.

Litterfall

We captured around 30 kg of litter on a combined littertrap surface of 30.24 ± 0.01 m² between May 17th 2016 and October fourth 2017. This translated into the production of 9.8 ± 0.2 Mg of leaf litter per hectare of forest over the study period, or 6.5 ± 0.2 Mg ha^{-1} y^{-1} during one year (from September 23rd 2016 until September 20th 2017). Litterfall showed large spatial variation even within blocks, leading to within-block CVs ranging between 17 and 139%, with an average CV of 55 and a median of 52%. Litterfall was greater during the months of June, July, August, September and October compared with the rest of the year (Fig 4). Overall, litterfall was highest in August (0.75 Mg ha $month^{-1}$) and lowest in November (0.42 Mg ha $month^{-1}$).

The carbon (C) content of the intercepted litter was, on average, 50.8 ± 0.1% over the entire period. The concentrations of N and P in the leaf litter averaged at 12.1 ± 0.1 g kg^{-1} and 0.224 ± 0.008 g kg^{-1}. The average weight-based and molar C:N ratios were 42.0 ± 0.4 and 49.0 ± 0.5, respectively, while the average weight-based and molar N:P ratios were 54 ± 2 and 119 ± 5, respectively. The concentrations of K, Ca and Mg fell between the N and P concentrations, with 1.6 ± 0.1 g kg^{-1}, 6.42 ± 0.07 g kg^{-1} and 2.44 ± 0.04 g kg^{-1}, respectively (Table 3). With the exception of K, which was roughly equal in bulk precipitation and three times higher in throughfall, the yearly deposition of each of the above measured macro-nutrients through litterfall was greater than its deposition by rain or throughfall. Compared to litterfall, the bulk precipitation and dry deposition together represented a large input of nutrients to the forest, with the exception of TDN, amounting to over 30% of the litterfall nutrient input for Ca, Mg

337 and P and over 100% for K. For throughfall these contributions were similar, with the
338 exception of K, which showed a throughfall input over three times larger than litterfall.

339 Discussion

340 The concentrations of most of the nutrients were negatively correlated to the deposition
341 volume while the fluxes were positively correlated (Table 2), indicating that when rainfall is
342 higher, solutes are more diluted. Interestingly, this was not the case for DOP which could
343 indicate that its deposition was unrelated to rainfall, but rather to deposition of biogenic
344 particles (including pollen, spores, bacteria, fungi, and small leaf fragments). Over the entire
345 study period, canopy interception was 25% of bulk precipitation volume and throughfall
346 volume correlated well with precipitation volume (Fig 3a). Canopy interception generally
347 ranges between 15 and 28% (Ponette-González et al. 2016), but smaller interception values
348 have been reported for neotropical forests and attributed to extremely high rainfall,
349 exceeding 4500 mm y^{-1} (Hofhansl et al. 2011). Similar to our findings for bulk precipitation
350 nutrient concentration in throughfall was also negatively correlated with throughfall volume.

351 The overall litterfall of 6.5 Mg ha^{-1} y^{-1} found in this study was similar to the mean South
352 American tropical forest annual leaf only litterfall of 6.1 Mg ha^{-1} y^{-1} reported by Chave et al.
353 (2010), who aggregated results from 52 old-growth forest sites, and leaf litterfall values
354 reported specifically for the Paracou research station (Wagner et al. 2013). We observed an
355 increase in litterfall at the beginning of the 2016 dry season, but in 2017 the litterfall increase
356 started a month earlier, already in June (Fig 4). Litterfall was captured between May 2016 and
357 September 2017, which is too short to draw any firm conclusions on litterfall seasonality, but
358 typically, litterfall is higher at the onset of the dry season which is attributed to seasonality in
359 solar radiation and rainfall (Myneni et al. 2007). Litterfall in Paracou was previously shown to

be seasonal (Chave et al. 2010) and litterfall is typically highest between July and October (Wagner et al. 2013; Wagner et al. 2016), which is the end of the wet season and onset of the dry season.

Nitrogen

The amount of TDN deposited by bulk precipitation was within the range reported for primary tropical forests situated in Central and South America (Boy et al. 2008; Hofhansl et al. 2011; Wilcke et al. 2013). Bulk precipitation and dry deposition flux of N_{in} amounted to ± 2 kg ha^{-1} y^{-1}, placing our study site into the lower range of N_{in} deposition values reported for lowland tropical forests, including a remote Central Amazonian site (~3 kg N_{in} ha^{-1} y^{-1}) (Pauliquevis et al. 2012), and within the range of 1 – 2 kg ha^{-1} y^{-1} reactive N deposition modelled by Wang et al. (2017) for 1997 until 2013. Anthropogenic sources of reactive N, such as fossil fuel burning, livestock operations and agriculture (Dämmgen and Erisman 2005), are scarce in French Guiana (Prospero et al. 2014), which may explain the low observed rates of N_{in} deposition compared to other tropical sites in Central America (Hofhansl et al. 2011), Africa (Galy-Lacaux and Delon 2014) or even Central Amazon (Andreae et al. 1990; Williams et al. 1997). There, often substantially higher rates of reactive N deposition were measured in tropical sites situated within 20 to 100 km of a major city or industry.

In contrast to our low N_{in}, the amount of DON in bulk precipitation fell at the high end of existing values reported for several South and Central American tropical forests (Boy et al. 2008; Hofhansl et al. 2011). However, DON values are not routinely measured and thus rarely reported, resulting in a specific knowledge gap for tropical forests (Jia et al. 2016). A possible source of DON at our site could be Amazon biomass burning, but the prevailing north-east wind direction in French Guiana (Aguilos et al. 2019) renders a substantial contribution of this N source unlikely. Instead, it is possible that at least a part of the deposited DON originated in

Africa and was transported across the Atlantic Ocean by the trade winds (Prospero et al. 1981). The intertropical convergence zone draws in aerosols from biomass burning in the African savannahs to the central Congo Basin (Bauters et al. 2018) resulting in extremely high DON deposition (Bauters et al. 2019). Because DON N can be transported by wind over distances of hundreds and thousands of kilometres (Cornell et al. 2003) the trade winds could carry aerosols from Savannah biomass burning containing different forms of organic N across the Atlantic Ocean (Jickells et al. 2016). There the DON is subsequently deposited, with highest deposition fluxes near the tropical Atlantic coast (Mahowald et al. 2005).

We found that there was no significant canopy exchange of NH_4^+, DON or TDN. Only NO_3^- was retained in the canopy at nearly 0.9 kg ha^{-1} y^{-1} (Table 3). The ability of forest canopies to retain N_{in} from atmospheric deposition is well known and believed to be important for tree nutrient uptake because it bypasses competition with soil microorganisms and understory, as was shown for temperate and boreal forests (Gaige et al. 2007; Sparks 2009). The magnitude of canopy uptake has been shown to depend on N deposition, forest N status and stand density in European forests (Schwarz et al. 2014), but we could find no such study carried out in the tropics. In tropical forests that are typically considered N rich (Hedin et al. 2009) and display no signs of N limitation, such as low N fixation rates (Van Langenhove et al. 2019) and high N export (Bauters et al. 2019), modest canopy uptake of N_{in} is unlikely to be of paramount significance to the trees N stocks. It may, however, contribute to growth of epiphytic lichens or bryophytes and to canopy dwelling bacteria which are all decoupled from the soil N cycle.

Phosphorus

We found modest TDP deposition that amounted to 0.5 kg ha^{-1} y^{-1} for bulk and dry deposition combined (Fig 5), which is similar to the reported average P deposition (0.43 kg ha^{-1} y^{-1}) for South and Central America combined (Tipping et al. 2014). Roughly 80% of the

deposited TDP was P_{in}, which is much higher than the global average 40% P_{in} contribution to TDP (Tipping et al. 2014). This suggests that the contribution of P derived from biogenic particles (including pollen, spores, bacteria, fungi, and fragments of leaves, which contain P in predominantly organic form) was generally lower than the global average. Instead, this ratio is indicative of dust deposition as soluble P in dust is primarily found in inorganic form (Graham et al. 2003). Typically, soil dust particles originate from local sources within the forest, but moist soils just like the soils at our site are less susceptible to wind erosion. Another likely source of P containing dust are the trade winds, which carry soil dust mostly originating from the Bodélé Depression in northern Chad (Ben-Ami et al. 2010; Koren et al. 2006; Swap et al. 1992). This dust contains, among other elements, P (Zamora et al. 2013) and has been proposed as a major nutrient input into the Amazon (Yu et al. 2015). A recent study calculated that, on average, both African biomass burning and dust transported across the Atlantic Ocean provides 80 g P ha^{-1} y^{-1} into the Amazon (Barkley et al. 2019), but this value is averaged for the entire Amazon basin and is likely higher in French Guiana than deeper in the continent (Prospero et al. 2014; Yu et al. 2015). A recent study in Panama even found that in periods with highest Saharan dust deposition up to 88 ± 31 g P ha^{-1} $month^{-1}$ arrives from Africa (Gross et al. 2016).

We found significant canopy uptake of TDP, which was primarily due to a slight P_{in} uptake of ± 150 g ha^{-1} y^{-1} (Table 3, Fig 5). This contrasts with other studies in tropical forests where authors typically find canopy leaching of P_{in} at varying rates between 0.1 and 2.2 kg ha^{-1} y^{-1} (Hofhansl et al. 2011; Tobón et al. 2004), which is attributed to high litter P contents, animal defecation in the canopy and leaching of decomposing canopy material. Tobón et al. (2004), who looked at throughfall fluxes in several tropical forests of Colombia, found that forest with lower soil P availabilities released less P_{in} from their canopies and sometimes even took up P_{in}.

Researchers reached a similar conclusion in a mature tropical dry forest in Mexico (Runyan et al. 2013) and together this suggests that when the forest P economy is tight less P is leached. Based on stoichiometric analyses, the forests of French Guiana seem severely P limited (Grau et al. 2017) and P added to the forest soil is immediately taken up by plant roots, indicating that P cycling is fast and efficient in this forest (Van Langenhove et al. 2020). Compared to other tropical rainforests (Wood et al. 2006) we found low litterfall P concentrations and very high N:P ratios, which can be interpreted as an indication of P limitation (Koerselman and Meuleman 1996). Because of our three week time interval between littertrap samplings, however, we cannot exclude leaching losses that may have slightly altered the N:P ratio before analysis. Although not as accurate as foliar N:P ratios, leaf litter N:P ratios have been used to assess plant nutrient limitation (Alvarez-Clare and Mack 2015), as larger values are indicative of P conservation by resorption before leaf abscission, resulting in lower element concentrations in litterfall (Vitousek 1984). Under these conditions indicative of a tight P economy, it seems plausible that canopy dwelling bacteria and epiphytes would profit from the deposited P_{in} for their metabolism and cause net uptake of deposited P_{in} compared to throughfall, just as was previously shown to occur for N (Umana and Wanek 2010).

On the ecosystem scale, the P input to the forest floor by litterfall is low at only 1.5 kg ha^{-1} y^{-1} and, compared to the input of external P by atmospheric deposition, is equivalent to roughly one third of the litterfall P (Figure 5). Relatively, the external input of atmospheric P compared to the internal recycling by litterfall P was similar to what was found in Ecuador (Wilcke et al. 2019), but higher than in other tropical forests in Panama (Gross et al. 2016) or Costa Rica (Hofhansl et al. 2011). In Panama Saharan dust deposition atmospheric P input was equivalent to between 10 and 29% of the P in monthly litterfall while in Costa Rica atmospheric P input amounted to 12% of the litterfall P input. However in Costa Rica, the authors

456 considered their forest sites less P limited than previously thought, due to the low P use
457 efficiencies and relatively high litter P content (roughly 0.6 g P kg^{-1}). Global estimates revealed
458 that the importance of atmospheric P input was greatest in locations with highest inputs, such
459 as deserts, and places where soil P is lowest, such as the Amazon Basin (Okin et al. 2004). In
460 our forest site both soil P stocks and extractable P are low (Courtois et al. 2018; Sabatier et al.
461 1997), litterfall N:P ratios are high, and additional P added to the soil is rapidly taken up by the
462 plant root system (Van Langenhove et al. 2020). Taken together, this seems to indicate if not
463 P limitation, at least a tight P cycling. Thus, just as was found in Ecuador (Wilcke et al. 2019),
464 the P input from the atmosphere likely represents a relevant external source of P for the
465 ecosystem.

466 Potassium

467 Potassium is the most abundant cation in plant cells and is the second most abundant
468 nutrient in leaves. However, it has often been neglected in studies of biogeochemistry and
469 stoichiometry in terrestrial ecosystems (Sardans and Peñuelas 2015). In this study, the
470 deposition flux of K^+ amounted to 14 kg ha^{-1} y^{-1} (Table 3), which was within the range reported
471 for other tropical sites (Boy et al. 2008), and less than 20% of this deposition was of marine
472 origin (Fig 2). Non-marine sources of K^+ include soil dust, K-containing fertilizers, biogenic (e.g.
473 spores, pollen and plant parts) and anthropogenic aerosols, and biomass burning (Berner and
474 Berner 2012). Given the low industrial and agricultural activities in French Guiana (Prospero
475 et al. 2014) biogenic aerosols and biomass burning are the most likely sources of K^+ there.
476 However, Saharan dust contains appreciable amounts of K (Moreno et al. 2006) and was
477 recently shown to deposit K as far as Manaus, Brazil (Rizzolo et al. 2017), thus it is not unlikely
478 that at least a part of the deposited K^+ originated in Africa.

The large amount of leached K^+ (Table 3) is in line with what is found in other forests around the world where K^+ deposition and subsequent movement through the canopy generally results in the net removal of K from canopy surfaces in all biomes (Ponette-González et al. 2016). Not only is dry-deposited K^+ highly soluble in water, but internal plant K^+ is concentrated in cells near leaf surfaces (Schlesinger and Bernhardt 2013) and thus sensitive to leakage following, e.g., cuticular damage. As a result, K^+ is easily washed from canopy surfaces and leached from plant tissues; thus K^+ leaching is generally high in wet environments such as tropical forests, sometimes reaching up to 147 kg K^+ leached $ha^{-}1\ y^{-1}$ (Boy and Wilcke 2008).

Here, we found that K cycling through litterfall was lower than in other tropical sites (Hofhansl et al. 2011; Wood et al. 2006) and only amounted to 10.5 kg $ha^{-1}\ y^{-1}$, which was lower than the combined bulk and dry deposition (14 kg $ha^{-1}\ y^{-1}$, Table 3). It is unknown if K is limiting in our study site, but the low litter input rates seem to suggest it might be, and if this is indeed the case the more than doubling of soil K input through the atmospheric deposition (Figure 5) represents an important K source to the ecosystem.

Chloride

Both Cl^- and Na^+ are the most abundant ions in seawater, between them constituting ± 85% of the total salinity in the oceans, and it is commonly accepted that the primary source of atmospheric chloride comes from the ocean. In areas close to the sea the Na^+/Cl^- ratio in precipitation is typically that of sea salt (Keene et al. 1986), thus the sea salt fraction of Cl^- in these coastal areas is typically close to 100%. This was not observed at our site where Cl^- deposition fluxes far exceeded those of Na^+ and the sea salt fraction of Cl^- was just below 50%, which is highly surprising for a forest situated close to the Ocean. French Guiana has few of the typical anthropogenic Cl^- sources, such as industrial and public fossil fuel combustion

503 (Prospero et al. 2014), and no volcanic emissions that can cause large Cl^- inputs (Aiuppa et al.
504 2006), so these are unable to explain the unexpectedly high Cl^- deposition. However, about 15
505 km away from the study site is the Guiana Space Centre (CSG), the main French and European
506 spaceport where during our measuring period twelve carrier rockets were launched. Each
507 carrier rocket had two booster rockets fuelled by 135 tons of a mixture of ammonium
508 perchlorate (68%), aluminium fuel (18%) and hydroxyl-terminated polybutadiene (14%) (ESA
509 2005). The combustion of ammonium perchlorate results in the production of water,
510 dioxygen, dinitrogen and hydrogen chloride. Hydrogen chloride is found primarily in gaseous
511 form at room temperatures and above (Breuer 2002) and can thus be deposited over a large
512 area in the days and weeks following the launch, as was shown at the John F. Kennedy space
513 centre, USA (Dreschel and Hall 1990). This may explain the high non-sea-salt fraction and
514 deposition of Cl^-, which is more than five times higher than observed at other tropical sites,
515 many of which are situated near the coast (Vet et al. 2014).

516 ## Calcium, magnesium and sulphate

517 The deposition flux of Mg^{2+} was within the ranges reported for other tropical sites, while
518 the amount of deposited Ca^{2+} and SO_4^{2-} was relatively high (Boy et al. 2008). The sea salt
519 contributions of Ca^{2+} and SO_4^{2-} were below 25%, while that for Mg^{2+} was above 80%,
520 illustrating that their main sources likely differed and that most of the Mg^{2+} was of marine
521 origin. The deposition of Ca^{2+} and SO_4^{2-} correlated well over time (data not shown), which
522 could indicate that both were deposited together, possibly as calcium sulphate ($CaSO_4$),
523 although we know of no local sources of $CaSO_4$ emissions that could explain its relatively high
524 deposition. Boy and Wilcke (2008) found that deposition of Ca^{2+} in an Andean forest was
525 mainly derived from Sahara dust and during their measurement period it exceeded 15 kg ha^{-1}
526 y^{-1}, higher than in many other tropical sites (Boy et al. 2008). Researchers calculated that for

527 Barbados, which just like French Guiana sits in the path of the transatlantic dust transported
528 from Africa, only 40% of the deposited non-sea-salt sulphate fraction was derived locally and
529 hypothesized that 60% may have come from African dust (Savoie et al. 1989).

530 We observed net canopy uptake of Ca^{2+} and SO_4^{2-}, while Mg^{2+} was not retained in or
531 leached from the canopy. For Ca^{2+} and SO_4^{2-}, this is unlike most other studies carried out in
532 the neotropics where mostly leaching of Ca^{2+} (Boy and Wilcke 2008) and SO_4^{2-} (Junior et al.
533 2015) was found, but no canopy exchange has also been reported for SO_4^{2-} (Zhang et al. 2007).
534 One of the reasons that in this study the canopy uptake of both these ions is so high is because,
535 according to the canopy budget model, the dry deposition is unusually high (Table 3). This
536 model makes two important assumptions that could lead to inaccuracies. Firstly it assumes
537 that all ions are deposited with equal efficiency as Na^+, which is not the case (Ruijgrok et al.
538 1997), and secondly it assumes that Na^+ is an inert tracer ion that is not influenced by canopy
539 exchange, which is also not always true (Staelens et al. 2008; Wanek et al. 2007). However, in
540 the case of SO_4^{2-} and Ca^{2+}, even disregarding the contribution of dry deposition would still lead
541 to a net canopy uptake of these ions, which is uncommon. One notable exception is a study
542 from two forests in southern Venezuela (Jordan et al. 1980). There, the authors found that
543 SO_4^{2-} and Ca^{2+} throughfall flux was much lower than precipitation flux and hypothesized this
544 was a nutrient conservation strategy of their forests after it had been predicted that these
545 nutrients were limiting in the region. We have no knowledge of SO_4^{2-} or Ca^{2+} nutrient scarcity
546 in our study site, but canopy epiphytes may represent one source of canopy uptake as several
547 studies have shown that they can take up nutrients including SO_4^{2-} and Ca^{2+} from the incoming
548 rainfall (Van Stan and Pypker 2015).

Conclusion

We found low bulk precipitation rates of N_{in}, while DON deposition was relatively high. Low rates of N_{in} deposition are consistent with low anthropogenic emissions, while high DON could be derived from biomass burning, either in the nearby Amazon or, at least to some extent, transported from Africa by the trade winds. Bulk precipitation P deposition was within the range of deposition rates reported for other tropical forest, but instead of P leaching from the canopy we found significant canopy uptake of P_{in}, which could be due to retention by canopy bryophytes, epiphytes and canopy dwelling bacteria. The yearly bulk atmospheric P deposition represented 30% of the annual P input to the forest soil by litterfall and the high litterfall N:P ratios were likely because of efficient P resorption, an indication of tight P cycling. Atmospheric P deposition, which here represented 20% of the annual P input to the forest floor (wet deposition, dry deposition and litterfall) after accounting for canopy uptake of P_{in}, is thus likely important for replenishing the P lost through erosion and soil leaching in this, arguably, P limited tropical forest.

The K^+ bulk precipitation flux was similar to other tropical sites, however the litterfall showed surprisingly low K^+ concentrations, amounting to less K^+ input to the forest floor than through bulk precipitation. There is likely efficient resorption of foliar K^+ before abscission, but the severe canopy leaching shows that throughfall provides an important pathway of K^+ input to the soil and trees with efficient soluble K^+ uptake mechanisms could greatly benefit from this input.

Lastly, the deposition rate of Cl^- was high and a large percentage of this flux could not be linked to sea salt depositions, which is typical for study sites near the coast. We suspect that the nearby activities of the European spaceport, and specifically their ammonium

perchlorate fuelled rocket launches, have caused large inputs of Cl^- that could not be linked to marine depositions.

Acknowledgements

This research was supported by the European Research Council Synergy grant ERC-2013-SyG.610028-IMBALANCE-P and the Paracou station is supported by an “Investissement d’Avenir” grant from the Agence Nationale de la Recherche (CEBA, ref ANR-10-LABX-25-01). We are grateful to Benoit Burban for providing us with the rainfall data collected at the Paracou flux tower, to Valentine Alt for assistance in the field and for Tom Van der Spiet for laboratory analyses.

Tables

Table 1 The range, mean and median of coefficients of variation (CVs) for the ions measured in the bulk deposition. For each sampling period a CV was calculated (N = 3).

	NO_3^--N	NH_4^+-N	DON	TDN	P_{in}	DOP	TDP	Na^+	Cl^-	K^+	Ca^{2+}	Mg^{2+}	SO_4^{2-}
Min CV (%)	1	8	3	2	3	16	3	1	2	1	1	2	0
Max CV (%)	88	173	137	105	173	173	161	87	60	102	87	173	87
Mean CV (%)	30	77	30	26	53	119	43	23	14	36	17	37	16
Median CV (%)	26	67	23	20	39	173	28	16	11	32	14	28	3

Table 2 Pearson correlations between the captured rainfall volume (as bulk precipitation and throughfall) and the concentration (mg L^{-1}) and deposition flux (kg ha^{-1} y^{-1}) of each ion.

	Bulk Precipitation		Throughfall	
	Concentration	Flux	Concentration	Flux
NO_3^--N	-0.280	0.559***	-0.483***	0.655***
NH_4^+-N	-0.438**	0.442**	-0.544***	0.378*
DON	-0.445**	0.691***	-0.587***	0.754***
TDN	-0.487***	0.736***	-0.592***	0.769***
P_{in}	-0.350*	0.489**	-0.490***	0.398*
DOP	-0.057	0.247	-0.496***	0.285
TDP	-0.372**	0.531***	-0.573***	0.403**
Na^+	-0.221	0.550***	-0.493***	0.685***
Cl^-	-0.208	0.754***	-0.376**	0.780***
SO_4^{2-}	-0.093	0.777***	-0.569***	0.839***
K^+	-0.088	0.736***	-0.489***	0.605***
Ca^{2+}	-0.461**	0.818***	-0.612***	0.846***
Mg^{2+}	-0.392**	0.584***	-0.630***	0.661***

Table 3 The average concentrations of each ion (mg L^{-1}) over the entire study period (Aug 2016 – Nov 2017) and for one calendar year (September 2016 until September 2017) in both bulk deposition and throughfall. The average yearly ion fluxes (kg ha^{-1} y^{-1}) by bulk deposition, throughfall, dry deposition and canopy exchange are also listed. Positive canopy exchange values indicate canopy leaching while negative values indicate canopy uptake. Element flux through litterfall (kg ha^{-1} y^{-1})is given for K, Ca, Mg, P, N and C for the period 23/09/2016 until 20/09/2017. Values in parentheses represent standard errors. Significant canopy exchanges (obtained using the mixed models) are given by * ($p < 0.05$), ** ($p < 0.01$) and *** ($p < 0.001$). Non-significant differences are indicated by NS. The listed N:P ratio is molar based. Values in parentheses are standard errors.

	NO_3^--N	NH_4^+-N	DON	TDN	PO_4^{2-}-P	DOP	TDP	Na^+	Cl^-	K^+	Ca^{2+}	Mg^{2+}	SO_4^{2-}	C	N:P
Bulk deposition concentration over study period (mg L^{-1})	0.031 (0.005)	0.020 (0.004)	0.25 (0.02)	0.30 (0.02)	0.010 (0.002)	0.002 (0.001)	0.013 (0.002)	0.93 (0.13)	3.38 (0.20)	0.32 (0.02)	0.78 (0.06)	0.16 (0.02)	1.43 (0.04)	-	51 (4)
Throughfall concentration over study period (mg L^{-1})	0.020 (0.002)	0.033 (0.010)	0.47 (0.04)	0.52 (0.05)	0.011 (0.002)	0.007 (0.002)	0.018 (0.004)	1.63 (0.18)	3.98 (0.40)	1.54 (0.25)	1.01 (0.107)	0.28 (0.03)	1.72 (0.08)	-	64 (7)
Bulk deposition concentration for one year (mg L^{-1})	0.031 (0.004)	0.019 (0.003)	0.25 (0.02)	0.30 (0.02)	0.009 (0.002)	0.002 (0.001)	0.011 (0.002)	0.95 (0.14)	3.43 (0.19)	0.36 (0.02)	0.77 (0.05)	0.16 (0.02)	1.41 (0.05)	-	60 (4)
Throughfall concentration for one year (mg L^{-1})	0.018 (0.002)	0.028 (0.008)	0.44 (0.03)	0.49 (0.04)	0.01 (0.002)	0.005 (0.001)	0.015 (0.003)	1.62 (0.20)	4.00 (0.45)	1.39 (0.25)	0.97 (0.07)	0.27 (0.03)	1.69 (0.08)	-	72 (6)
Bulk deposition (kg ha^{-1} y^{-1})	1.092 (0.161)	0.677 (0.119)	8.75 (0.62)	10.51 (0.73)	0.319 (0.054)	0.084 (0.024)	0.403 (0.036)	33.8 (4.6)	121.9 (7.0)	11.2 (0.7)	27.2 (1.8)	5.7 (0.6)	50.1 (1.9)	-	58 (5)
Dry deposition (kg ha^{-1} y^{-1})	0.275 (0.223)	0.167 (0.136)	2.20 (1.77)	2.64 (2.12)	0.080 (0.067)	0.021 (0.019)	0.101 (0.083)	8.5 (6.9)	30.6 (24.6)	2.8 (2.3)	6.9 (5.5)	1.4 (1.2)	12.6 (10.1)		58
Throughfall (kg ha^{-1} y^{-1})	0.482 (0.051)	0.743 (0.223)	11.44 (0.9)	12.67 (1.10)	0.255 (0.053)	0.136 (0.038)	0.395 (0.075)	42.5 (5.0)	104.6 (11.3)	36.5 (6.1)	25.3 (1.8)	7.0 (0.8)	44.2 (2.3)	-	71 (7)
Canopy exchange (kg ha^{-1} y^{-1})	-0.890 (0.270) ***	-0.108 (0.268) NS	0.37 (2.06) NS	-0.60 (2.48) NS	-0.144 (0.111) *	0.031 (0.048) NS	-0.107 (0.079) *	0 (0) NS	-48.8 (27.8)*	21.5 (6.4)** *	-9.1 (6.0)*	-0.2 (1.5) NS	-18.6 (10.5)* *		12
Litterfall (kg ha^{-1} y^{-1})	-	-	-	81.75 (3.00)	-	-	1.52 (0.08)	-	-	10.6 (0.5)	41.8 (1.6)	15.7 (0.6)	-	3292 (122)	119 (5)

Figures

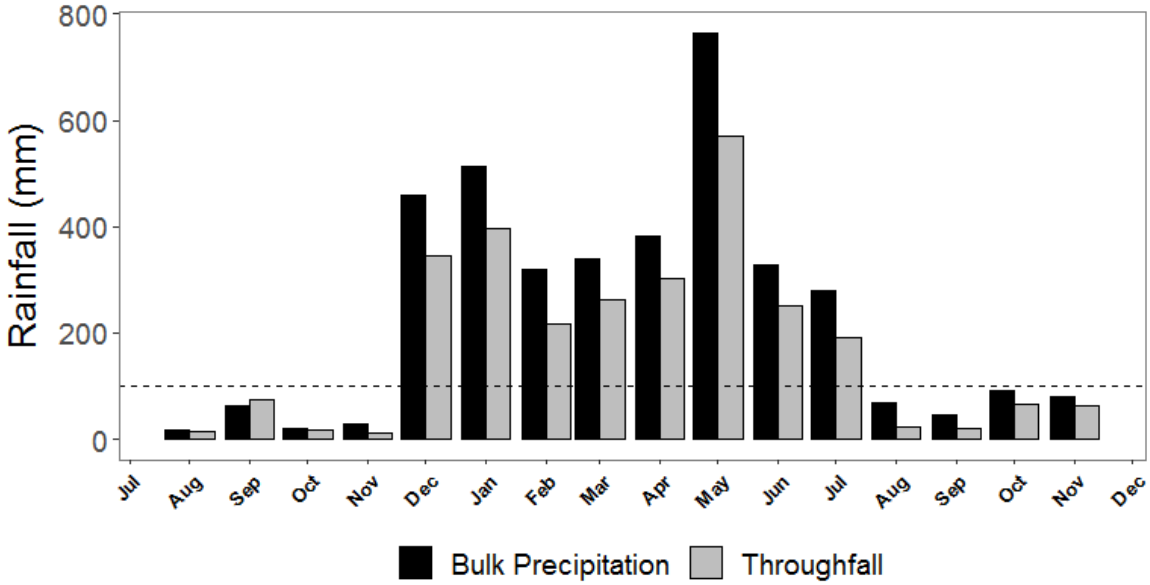

Figure 1 Mean monthly precipitation measured in the bulk precipitation (black) and throughfall (grey) over the period August 19th 2016 until November 15th 2017. The horizontal dashed line indicates a precipitation of 100 mm and all months with lower precipitation are considered dry season months

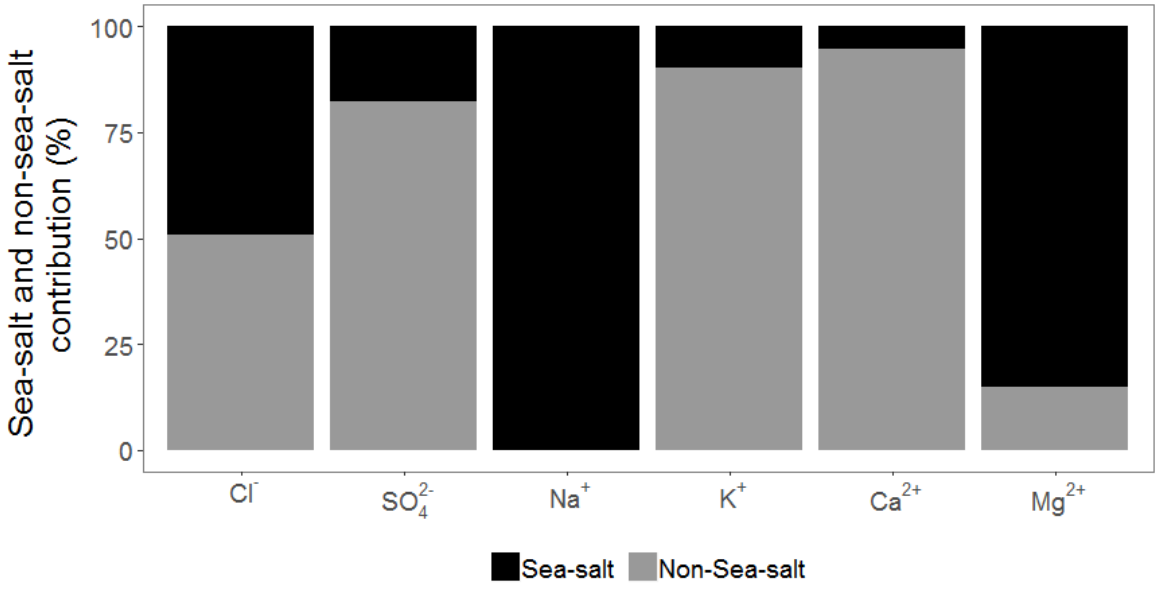

Figure 2 The marine (sea-salt, black) or non-marine (non-sea-salt, grey) origin of Cl^-, SO_4^{2-}, K^+, Ca^{2+} and Mg^{2+} expressed as a percentage, assuming all Na^+ was marine in origin (see Berner and Berner 2012).

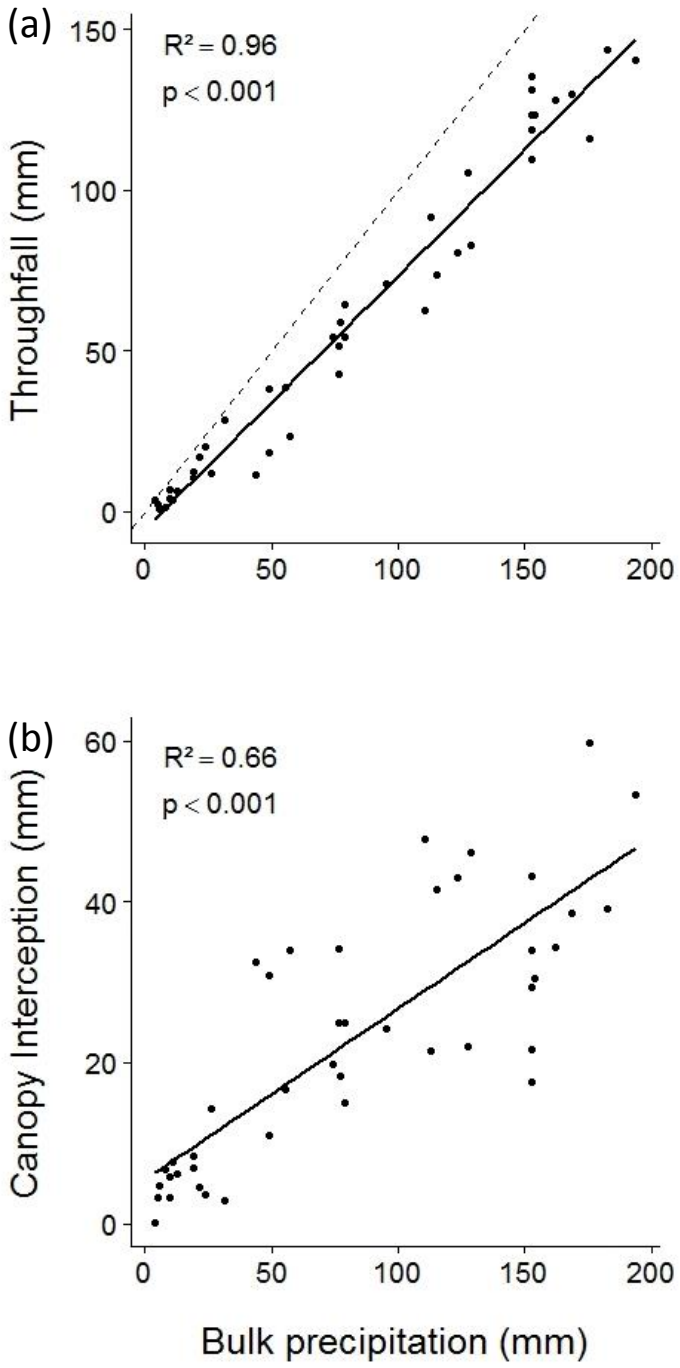

Figure 3 Relationship between (a) bulk precipitation and the throughfall volume measured for a given sampling period, and (b) bulk precipitation and the precipitation volume intercepted by the canopy. The 1:1 line in graph (a) is given by the dashed black line. Both the R^2 and the significance of the Pearson correlation are given.

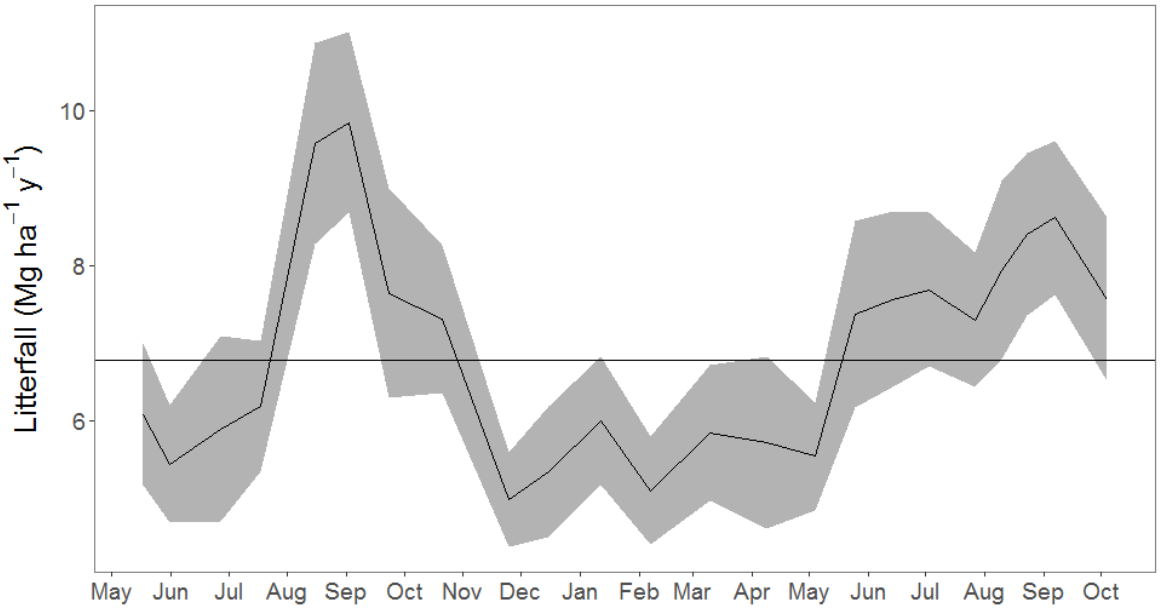

Figure 4 Litterfall over the entire study period. The 95% confidence interval is depicted by the grey band and the overall average litterfall is depicted by the horizontal line. Note that the y-axis does not start on 0.

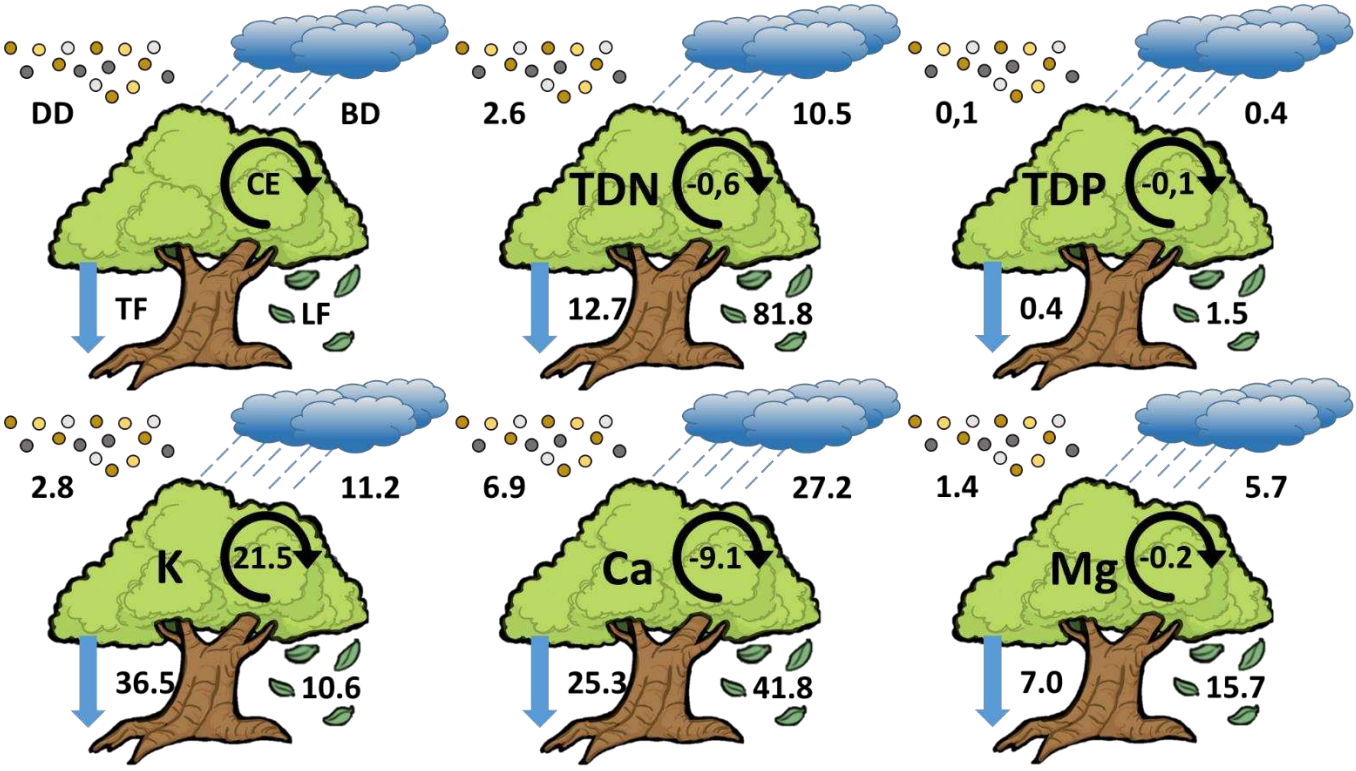

Figure 5 Overview of the inputs, outputs and canopy exchange measured in this study for total dissolved nitrogen (TDN), total dissolved phosphorus (TDP), potassium (K), calcium (Ca) and magnesium (Mg). Negative values for CE indicate canopy uptake and positive values indicate canopy leaching. Value units are kg ha^{-1} y^{-1}. BD = Bulk deposition, DD = Dry deposition, TF = Throughfall, CE = Canopy Exchange and LF = Litterfall

References

Aguilos M, Stahl C, Burban B et al. (2019) Interannual and Seasonal Variations in Ecosystem Transpiration and Water Use Efficiency in a Tropical Rainforest Forests 10:14

Aiuppa A, Bellomo S, Brusca L, D'Alessandro W, Di Paola R, Longo M (2006) Major-ion bulk deposition around an active volcano (Mt. Etna, Italy) Bulletin of Volcanology 68:255-265 doi:10.1007/s00445-005-0005-x

Alvarez-Clare S, Mack MC (2015) Do Foliar, Litter, and Root Nitrogen and Phosphorus Concentrations Reflect Nutrient Limitation in a Lowland Tropical Wet Forest? PloS one 10:e0123796 doi:10.1371/journal.pone.0123796

Andreae MO, Talbot RW, Berresheim H, Beecher KM (1990) Precipitation chemistry in central Amazonia Journal of Geophysical Research: Atmospheres 95:16987-16999 doi:10.1029/JD095iD10p16987

Baillie IC (1996) Soils of the humid tropics. In: Richards PW (ed) The tropical rain forest. Cambridge University Press, Cambridge, pp 256-285

Bales RC, Conklin MH, Kerkez B et al. (2011) Sampling Strategies in Forest Hydrology and Biogeochemistry. In: Levia DF, Carlyle-Moses D, Tanaka T (eds) Forest Hydrology and Biogeochemistry: Synthesis of Past Research and Future Directions. Springer Netherlands, Dordrecht, pp 29-44. doi:10.1007/978-94-007-1363-5_2

Barkley AE, Prospero JM, Mahowald N et al. (2019) African biomass burning is a substantial source of phosphorus deposition to the Amazon, Tropical Atlantic Ocean, and Southern Ocean Proceedings of the National Academy of Sciences 116:16216-16221 doi:10.1073/pnas.1906091116

Bauters M, Drake TW, Verbeeck H et al. (2018) High fire-derived nitrogen deposition on central African forests Proceedings of the National Academy of Sciences of the United States of America 115:549-554 doi:10.1073/pnas.1714597115

Bauters M, Verbeeck H, Rütting T et al. (2019) Contrasting nitrogen fluxes in African tropical forests of the Congo Basin Ecol Monogr 89:e01342 doi:doi:10.1002/ecm.1342

Ben-Ami Y, Koren I, Rudich Y, Artaxo P, Martin ST, Andreae MO (2010) Transport of North African dust from the Bodélé depression to the Amazon Basin: a case study Atmos Chem Phys 10:7533-7544 doi:10.5194/acp-10-7533-2010

Berner EK, Berner RA (2012) Global Environment Water, Air, and Geochemical Cycles. Second Edition edn. Princeton University Press, Princeton, USA

Bonal D, Bosc A, Ponton S et al. (2008) Impact of severe dry season on net ecosystem exchange in the Neotropical rainforest of French Guiana Global Change Biology 14:1917-1933 doi:10.1111/j.1365-2486.2008.01610.x

Boy J, Rollenbeck R, Valarezo C, Wilcke W (2008) Amazonian biomass burning-derived acid and nutrient deposition in the north Andean montane forest of Ecuador Global Biogeochem Cy 22:n/a-n/a doi:10.1029/2007gb003158

Boy J, Wilcke W (2008) Tropical Andean forest derives calcium and magnesium from Saharan dust Global Biogeochem Cy 22 doi:10.1029/2007gb002960

Bredemeier M (1988) Forest canopy transformation of atmospheric deposition Water, Air, and Soil Pollution 40:121-138 doi:10.1007/BF00279460

Breuer D (2002) Volatile Inorganic Acids (HCl, HBr, HNO3). In: Greim H, Kettrup A (eds) Analyses of Hazardous Substances in Air, vol Six. Wiley-VCH, pp 211-223. doi:10.1002/352760023X.ch16

Chave J, Navarrete D, Almeida S et al. (2010) Regional and seasonal patterns of litterfall in tropical South America Biogeosciences 7:43-55 doi:10.5194/bg-7-43-2010

Cornell SE, Jickells TD, Cape JN, Rowland AP, Duce RA (2003) Organic nitrogen deposition on land and coastal environments: a review of methods and data Atmospheric Environment 37:2173-2191 doi:https://doi.org/10.1016/S1352-2310(03)00133-X

Courtois EA, Stahl C, Van den Berge J et al. (2018) Spatial Variation of Soil CO2, CH4 and N2O Fluxes Across Topographical Positions in Tropical Forests of the Guiana Shield Ecosystems doi:10.1007/s10021-018-0232-6

Dämmgen U, Erisman JW (2005) Emission, transmission, deposition and environmental effects of ammonia from agricultural sources. In: Kuczunski T, Dämmgen U, Webb J, Myczko A (eds) Emissions from European agriculture. Wageningen Academic Publishers, Wageningen, pp 97 - 112

Deusdará KRL, Forti MC, Borma LS, Menezes RSC, Lima JRS, Ometto JPHB (2016) Rainwater chemistry and bulk atmospheric deposition in a tropical semiarid ecosystem: the Brazilian Caatinga Journal of Atmospheric Chemistry:1-15 doi:10.1007/s10874-016-9341-9

Draaijers GPJ, Erisman JW, Spranger T, Wyers GP (1996) The application of throughfall measurements for atmospheric deposition monitoring Atmospheric Environment 30:3349-3361 doi:Doi 10.1016/1352-2310(96)00030-1

Dreschel TW, Hall CR (1990) Quantification of hydrochloric acid and particulate deposition resulting from space shuttle launches at John F. Kennedy space center, Florida, USA Environmental Management 14:501 doi:10.1007/BF02394138

Epron D, Bosc A, Bonal D, Freycon V (2006) Spatial variation of soil respiration across a topographic gradient in a tropical rain forest in French Guiana J Trop Ecol 22:565-574 doi:10.1017/S0266467406003415

ESA (2005) Launch Vehicles space transportation: Boosters (EAP). European Space Agency. https://www.esa.int/Our_Activities/Space_Transportation/Launch_vehicles/Boosters_EAP. Accessed 26/08/2019 2019

FAO (1998) World Reference Base for Soil Resources vol 84. Rome

Gaige E, Dail DB, Hollinger DY et al. (2007) Changes in Canopy Processes Following Whole-Forest Canopy Nitrogen Fertilization of a Mature Spruce-Hemlock Forest Ecosystems 10:1133-1147 doi:10.1007/s10021-007-9081-4

Galloway JN, dentener FJ, Capone DG et al. (2004) Nitrogen Cycles:Past, Present, and Future Biogeochemistry 70

Galy-Lacaux C, Delon C (2014) Nitrogen emission and deposition budget in West and Central Africa Environmental Research Letters 9:125002 doi:10.1088/1748-9326/9/12/125002

Gourlet-Fleury S, Guehl JM, Laroussinie O (2004) Ecology and management of a neotropical rainforest. Lessons drawn from Paracou, a long-term experimental research site in French Guiana. Elsevier, Meppel, The Netherlands

Graham B, Guyon P, Maenhaut W et al. (2003) Composition and diurnal variability of the natural Amazonian aerosol Journal of Geophysical Research: Atmospheres 108 doi:10.1029/2003jd004049

Grau O, Penuelas J, Ferry B et al. (2017) Nutrient-cycling mechanisms other than the direct absorption from soil may control forest structure and dynamics in poor Amazonian soils Sci Rep 7:45017 doi:10.1038/srep45017

Gross A, Turner BL, Goren T, Berry A, Angert A (2016) Tracing the Sources of Atmospheric Phosphorus Deposition to a Tropical Rain Forest in Panama Using Stable Oxygen Isotopes Environmental Science & Technology 50:1147-1156 doi:10.1021/acs.est.5b04936

Hedin LO, Brookshire ENJ, Menge DNL, Barron AR (2009) The Nitrogen Paradox in Tropical Forest Ecosystems Annual Review of Ecology, Evolution, and Systematics 40:613-635 doi:10.1146/annurev.ecolsys.37.091305.110246

Hobbs PV (2000) Introduction to Atmospheric Chemistry. Cambridge University Press, Cambridge. doi:DOI: 10.1017/CBO9780511808913

Hofhansl F, Wanek W, Drage S, Huber W, Weissenhofer A, Richter A (2011) Topography strongly affects atmospheric deposition and canopy exchange processes in different types of wet lowland rainforest, Southwest Costa Rica Biogeochemistry 106:371-396 doi:10.1007/s10533-010-9517-3

Hofhansl F, Wanek W, Drage S, Huber W, Weissenhofer A, Richter A (2012) Controls of hydrochemical fluxes via stemflow in tropical lowland rainforests: Effects of meteorology and vegetation characteristics Journal of Hydrology 452-453:247-258 doi:10.1016/j.jhydrol.2012.05.057
Houlton BZ, Morford SL, Dahlgren RA (2018) Convergent evidence for widespread rock nitrogen sources in Earth's surface environment Science (New York, NY) 360:58-62 doi:10.1126/science.aan4399
Jia Y, Yu G, Gao Y, He N, Wang Q, Jiao C, Zuo Y (2016) Global inorganic nitrogen dry deposition inferred from ground- and space-based measurements Sci Rep 6:19810 doi:10.1038/srep19810
Jickells TD, Baker AR, Chance R (2016) Atmospheric transport of trace elements and nutrients to the oceans Philos Trans A Math Phys Eng Sci 374:20150286 doi:10.1098/rsta.2015.0286
Jordan C, Golley F, Hall J, Hall J (1980) Nutrient Scavenging of Rainfall by the Canopy of an Amazonian Rain Forest Biotropica 12:61-66 doi:10.2307/2387774
Junior RCdO, Keller MM, Ramos JFdF, Beldini TP, Crill PM, Camargo PBd, Haren Jv (2015) Chemical analysis of rainfall and throughfall in the Tapajós National Forest, Belterra, Pará, Brazil Revista Ambiente & Água 10:263-285
Keene WC, Pszenny AAP, Galloway JN, Hawley ME (1986) Sea-salt corrections and interpretation of constituent ratios in marine precipitation Journal of Geophysical Research: Atmospheres 91:6647-6658 doi:10.1029/JD091iD06p06647
Koerselman W, Meuleman AF (1996) The vegetation N: P ratio: a new tool to detect the nature of nutrient limitation Journal of applied Ecology:1441-1450
Koren I, Kaufman YJ, Washington R, Todd MC, Rudich Y, Martins JV, Rosenfeld D (2006) The Bodélé depression: a single spot in the Sahara that provides most of the mineral dust to the Amazon forest Environmental Research Letters 1:014005 doi:10.1088/1748-9326/1/1/014005
Kuznetsova A, Brockhoff PB, Christensen RHB (2017) lmerTest Package: Tests in Linear Mixed Effects Models Journal of Statistical Software 82:27
Mahowald NM, Artaxo P, Baker AR, Jickells TD, Okin GS, Randerson JT, Townsend AR (2005) Impacts of biomass burning emissions and land use change on Amazonian atmospheric phosphorus cycling and deposition Global Biogeochem Cy 19 doi:10.1029/2005gb002541
Moreno T, Querol X, Castillo S et al. (2006) Geochemical variations in aeolian mineral particles from the Sahara-Sahel Dust Corridor Chemosphere 65:261-270 doi:10.1016/j.chemosphere.2006.02.052
Mphepya JN, Pienaar JJ, Galy-Lacaux C, Held G, Turner CR (2004) Precipitation Chemistry in Semi-Arid Areas of Southern Africa: A Case Study of a Rural and an Industrial Site Journal of Atmospheric Chemistry 47:1-24 doi:10.1023/B:JOCH.0000012240.09119.c4
Myneni RB, Yang W, Nemani RR et al. (2007) Large seasonal swings in leaf area of Amazon rainforests Proceedings of the National Academy of Sciences 104:4820 doi:10.1073/pnas.0611338104
Okin GS, Mahowald N, Chadwick OA, Artaxo P (2004) Impact of desert dust on the biogeochemistry of phosphorus in terrestrial ecosystems Global Biogeochem Cy 18 doi:10.1029/2003gb002145
Pacyna JM (2008) Atmospheric Deposition. In: Jørgensen SE, Fath BD (eds) Encyclopedia of Ecology. Academic Press, Oxford, pp 275-285. doi:https://doi.org/10.1016/B978-008045405-4.00258-5
Pan Y, Birdsey RA, Fang J et al. (2011) A Large and Persistent Carbon Sink in the World's Forests Science (New York, NY) 333:988 doi:10.1126/science.1201609
Pauliquevis T, Lara LL, Antunes ML, Artaxo P (2012) Aerosol and precipitation chemistry measurements in a remote site in Central Amazonia: the role of biogenic contribution Atmospheric Chemistry and Physics 12:4987-5015 doi:10.5194/acp-12-4987-2012
Ponette-González AG, Ewing HA, Weathers KC (2016) Interactions between Precipitation and Vegetation Canopies. In: Johnson EA, Martin YE (eds) A Biogeoscience Approach to

Ecosystems. Cambridge University Press, Cambridge, pp 215-253. doi:DOI: 10.1017/CBO9781107110632.009
Porder S, Asner GP, Vitousek PM (2005) Ground-based and remotely sensed nutrient availability across a tropical landscape Proceedings of the National Academy of Sciences of the United States of America 102:10909-10912 doi:10.1073/pnas.0504929102
Prospero JM, Collard F-X, Molinié J, Jeannot A (2014) Characterizing the annual cycle of African dust transport to the Caribbean Basin and South America and its impact on the environment and air quality Global Biogeochem Cy 28:757-773 doi:10.1002/2013gb004802
Prospero JM, Glaccum RA, Nees RT (1981) Atmospheric transport of soil dust from Africa to South America Nature 289:570-572 doi:10.1038/289570a0
R Core Team (2018) R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing 3.5
Reed SC, Cleveland CC, Townsend AR (2007) Controls over leaf litter and soil nitrogen fixation in two lowland tropical rain forests Biotropica 39:585-592 doi:DOI 10.1111/j.1744-7429.2007.00310.x
Reed SC, Cleveland CC, Townsend AR (2011) Functional Ecology of Free-Living Nitrogen Fixation: A Contemporary Perspective Annu Rev Ecol Evol S 42:489-512 doi:DOI 10.1146/annurev-ecolsys-102710-145034
Rizzolo JA, Barbosa CGG, Borillo GC et al. (2017) Soluble iron nutrients in Saharan dust over the central Amazon rainforest Atmospheric Chemistry and Physics 17:2673-2687 doi:10.5194/acp-17-2673-2017
Ruijgrok W, Tieben H, Eisinga P (1997) The dry deposition of particles to a forest canopy: A comparison of model and experimental results Atmospheric Environment 31:399-415 doi:https://doi.org/10.1016/S1352-2310(96)00089-1
Runyan CW, Lawrence D, Vandecar KL, D'Odorico P (2013) Experimental evidence for limited leaching of phosphorus from canopy leaves in a tropical dry forest Ecohydrology 6:806-817 doi:10.1002/eco.1303
Sabatier D, Grimaldi M, Prévost M-F, Guillaume J, Godron M, Dosso M, Curmi P (1997) The influence of soil cover organization on the floristic and structural heterogeneity of a Guianan rain forest Plant Ecology 131:81-108 doi:10.1023/a:1009775025850
Sardans J, Peñuelas J (2015) Potassium: a neglected nutrient in global change Global Ecology and Biogeography 24:261-275 doi:10.1111/geb.12259
Savoie DL, Prospero JM, Saltzman ES (1989) Non-sea-salt sulfate and nitrate in trade wind aerosols at Barbados: Evidence for long-range transport Journal of Geophysical Research: Atmospheres 94:5069-5080 doi:10.1029/JD094iD04p05069
Schlesinger WH, Bernhardt ES (2013) Biogeochemistry: an Analysis of Global Change. 3rd edn. Academic Press, San Diego, CA
Schwarz MT, Bischoff S, Blaser S et al. (2014) More efficient aboveground nitrogen use in more diverse Central European forest canopies Forest Ecology and Management 313:274-282 doi:https://doi.org/10.1016/j.foreco.2013.11.021
See CR, Luke McCormack M, Hobbie SE, Flores-Moreno H, Silver WL, Kennedy PG (2019) Global patterns in fine root decomposition: climate, chemistry, mycorrhizal association and woodiness Ecology letters 22:946-953 doi:10.1111/ele.13248
Sparks JP (2009) Ecological ramifications of the direct foliar uptake of nitrogen Oecologia 159:1-13 doi:10.1007/s00442-008-1188-6
Staelens J, Houle D, De Schrijver A, Neirynck J, Verheyen K (2008) Calculating Dry Deposition and Canopy Exchange with the Canopy Budget Model: Review of Assumptions and Application to Two Deciduous Forests Water, Air, and Soil Pollution 191:149-169 doi:10.1007/s11270-008-9614-2
Sullivan BW, Smith WK, Townsend AR, Nasto MK, Reed SC, Chazdon RL, Cleveland CC (2014) Spatially robust estimates of biological nitrogen (N) fixation imply substantial human alteration of the

tropical N cycle Proceedings of the National Academy of Sciences of the United States of America 111:8101-8106 doi:10.1073/pnas.1320646111
Swap R, Garstang M, Greco S, Talbot R, Kållberg P (1992) Saharan dust in the Amazon Basin Tellus B: Chemical and Physical Meteorology 44:133-149 doi:10.3402/tellusb.v44i2.15434
Taylor BN, Chazdon RL, Menge DNL (2019) Successional dynamics of nitrogen fixation and forest growth in regenerating Costa Rican rainforests Ecology 100:e02637 doi:10.1002/ecy.2637
Taylor PG, Cleveland CC, Wieder WR, Sullivan BW, Doughty CE, Dobrowski SZ, Townsend AR (2017) Temperature and rainfall interact to control carbon cycling in tropical forests Ecology letters 20:779-788 doi:10.1111/ele.12765
Tipping E, Benham S, Boyle JF et al. (2014) Atmospheric deposition of phosphorus to land and freshwater Environmental Science: Processes & Impacts 16:1608-1617 doi:10.1039/C3EM00641G
Tobón C, Sevink J, Verstraten JM (2004) Solute fluxes in throughfall and stemflow in four forest ecosystems in northwest Amazonia Biogeochemistry 70:25 doi:https://doi.org/10.1023/B:BIOG.0000049334.10381.f8
Turner BL, Brenes-Arguedas T, Condit R (2018) Pervasive phosphorus limitation of tree species but not communities in tropical forests Nature 555:367-370 doi:10.1038/nature25789
Ulrich B (1983) Interaction of Forest Canopies with Atmospheric Constituents: So2, Alkali and Earth Alkali Cations and Chloride. In: Ulrich B, Pankrath J (eds) Effects of Accumulation of Air Pollutants in Forest Ecosystems: Proceedings of a Workshop held at Göttingen, West Germany, May 16–18, 1982. Springer Netherlands, Dordrecht, pp 33-45. doi:10.1007/978-94-009-6983-4_2
Umana NH-N, Wanek W (2010) Large Canopy Exchange Fluxes of Inorganic and Organic Nitrogen and Preferential Retention of Nitrogen by Epiphytes in a Tropical Lowland Rainforest Ecosystems 13:367-381 doi:10.1007/s10021-010-9324-7
Van Langenhove L, Depaepe T, Vicca S et al. (2019) Regulation of nitrogen fixation from free-living organisms in soil and leaf litter of two tropical forests of the Guiana shield Plant and Soil doi:10.1007/s11104-019-04012-1
Van Langenhove L, Janssens IA, Verryckt L et al. (2020) Rapid root assimilation of added phosphorus in a lowland tropical rainforest of French Guiana Soil Biology and Biochemistry 140:107646 doi:10.1016/j.soilbio.2019.107646
Van Stan JT, Pypker TG (2015) A review and evaluation of forest canopy epiphyte roles in the partitioning and chemical alteration of precipitation Science of The Total Environment 536:813-824 doi:https://doi.org/10.1016/j.scitotenv.2015.07.134
Vet R, Artz RS, Carou S et al. (2014) A global assessment of precipitation chemistry and deposition of sulfur, nitrogen, sea salt, base cations, organic acids, acidity and pH, and phosphorus Atmospheric Environment 93:3-100 doi:10.1016/j.atmosenv.2013.10.060
Vitousek PM (1984) Litterfall, Nutrient Cycling, and Nutrient Limitation in Tropical Forests Ecology 65:285-298 doi:10.2307/1939481
Vitousek PM, Porder S, Houlton BZ, Chadwick OA (2010) Terrestrial phosphorus limitation: mechanisms, implications, and nitrogen–phosphorus interactions Ecol Appl 20:5-15 doi:10.1890/08-0127.1
Wagner F, Rossi V, Stahl C, Bonal D, Hérault B (2013) Asynchronism in leaf and wood production in tropical forests: a study combining satellite and ground-based measurements Biogeosciences 10:7307-7321 doi:10.5194/bg-10-7307-2013
Wagner FH, Hérault B, Bonal D et al. (2016) Climate seasonality limits leaf carbon assimilation and wood productivity in tropical forests Biogeosciences 13:2537-2562 doi:10.5194/bg-13-2537-2016
Walinga I, Van Der Lee JJ, Houva VJG, Van Vark W, Novozamsky I (1995) Digestion in tubes with H2S04-salicylic acid, H202 and selenium and determination of Ca, K, Mg, N, Na, P, Zn. In: Walinga I, Van Der Lee JJ, Houva VJG, Van Vark W, Novozamsky I (eds) Plant Analysis Manual. Springer, Dordrecht, NL. doi:https://doi.org/10.1007/978-94-011-0203-2

Walker TW, Syers JK (1976) The fate of phosphorus during pedogenesis Geoderma 15:1-19 doi:https://doi.org/10.1016/0016-7061(76)90066-5
Wanek W, Hofmann J, Feller IC (2007) Canopy interactions of rainfall in an off-shore mangrove ecosystem dominated by Rhizophora mangle (Belize) Journal of Hydrology 345:70-79 doi:https://doi.org/10.1016/j.jhydrol.2007.07.012
Wang R, Goll D, Balkanski Y et al. (2017) Global forest carbon uptake due to nitrogen and phosphorus deposition from 1850 to 2100 Glob Chang Biol 23:4854-4872 doi:10.1111/gcb.13766
Whitehead HC, Feth JH (1964) Chemical composition of rain, dry fallout, and bulk precipitation at Menlo Park, California, 1957–1959 Journal of Geophysical Research (1896-1977) 69:3319-3333 doi:10.1029/JZ069i016p03319
Wickham H (2016) ggplot2: Elegant graphics for Data Analysis. Springer-Verlag, New York
Wieder WR, Cleveland CC, Smith WK, Todd-Brown K (2015) Future productivity and carbon storage limited by terrestrial nutrient availability Nat Geosci 8:441-444 doi:10.1038/ngeo2413
Wilcke W, Leimer S, Peters T et al. (2013) The nitrogen cycle of tropical montane forest in Ecuador turns inorganic under environmental change Global Biogeochem Cy 27:1194-1204 doi:10.1002/2012GB004471
Wilcke W, Velescu A, Leimer S, Bigalke M, Boy J, Valarezo C (2019) Temporal Trends of Phosphorus Cycling in a Tropical Montane Forest in Ecuador During 14 Years Journal of Geophysical Research: Biogeosciences doi:10.1029/2018jg004942
Williams MR, Fisher TR, Melack JM (1997) Chemical composition and deposition of rain in the central Amazon, Brazil Atmospheric Environment 31:207-217 doi:https://doi.org/10.1016/1352-2310(96)00166-5
Wood TE, Lawrence D, Clark DA (2006) Determinants of Leaf Litter Nutrient Cycling in a Tropical Rain Forest: Soil Fertility Versus Topography Ecosystems 9:700-710 doi:10.1007/s10021-005-0016-7
Wright SJ, Yavitt JB, Wurzburger N et al. (2011) Potassium, phosphorus, or nitrogen limit root allocation, tree growth, or litter production in a lowland tropical forest Ecology 92:1616-1625 doi:10.1890/10-1558.1
Wurzburger N, Bellenger JP, Kraepiel AM, Hedin LO (2012) Molybdenum and phosphorus interact to constrain asymbiotic nitrogen fixation in tropical forests PloS one 7:e33710 doi:10.1371/journal.pone.0033710
Xiao H-W, Xiao H-Y, Long A-M, Wang Y-L, Liu C-Q (2013) Chemical composition and source apportionment of rainwater at Guiyang, SW China Journal of Atmospheric Chemistry 70:269-281 doi:10.1007/s10874-013-9268-3
Yu HB, Chin M, Yuan TL et al. (2015) The fertilizing role of African dust in the Amazon rainforest: A first multiyear assessment based on data from Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations Geophysical Research Letters 42:1984-1991 doi:10.1002/2015gl063040
Zamora LM, Prospero JM, Hansell DA, Trapp JM (2013) Atmospheric P deposition to the subtropical North Atlantic: sources, properties, and relationship to N deposition Journal of Geophysical Research: Atmospheres 118:1546-1562 doi:10.1002/jgrd.50187
Zhang G, Zeng G-M, Du C-Y et al. (2007) Deposition patterns in bulk precipitation and throughfall in a subtropical mixed forest in central-south China Forestry: An International Journal of Forest Research 80:211-221 doi:10.1093/forestry/cpm002