

Title

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

Authors

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1 Abstract

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

16 Keywords

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

18 Introduction

19 Tropical forests are of major significance to the global carbon budget as they account for 70%
20 of the global gross forest carbon sink and about 55% of the global forest carbon pool is stored
21 in tropical forests (Pan et al. 2011). In order to sustain the high productivity and carbon sink-
22 strength of tropical forests, nutrient inputs are needed to compensate for the continuous
23 nutrient losses by biomass removal, leaching, erosion, or denitrification (Vitousek 1984;
24 Wieder et al. 2015). Three main natural pathways add nutrients to ecosystems, namely
25 mineral weathering, Nitrogen (N) fixation (only for N) and atmospheric deposition. Over short
26 timescales, weathering rates are thought to be slow in lowland tropical forests (Porder et al.
27 2005) as they are typically characterized by already highly weathered soils and little altitudinal
28 variation, which leads to slow rates of soil rejuvenation through erosion (Baillie 1996; Vitousek
29 et al. 2010; Walker and Syers 1976). In mature lowland tropical forests the input of external N
30 through free-living (soil, leaf litter) or symbiotic N fixation is highly variable and both high and
31 low rates have been reported (Reed et al. 2007; Reed et al. 2011; Sullivan et al. 2014; Taylor
32 et al. 2019; Van Langenhove et al. 2019; Wurzburger et al. 2012). Possibly, atmospheric
33 deposition represents an important external nutrient input for these tropical forests where
34 other external inputs, such as rock weathering (Houlton et al. 2018) or N fixation, are low
35 (Pacyna 2008) and could therefore be a key component of their nutrient budget (Hofhansl et
36 al. 2012). Unfortunately, there is a particular gap in atmospheric deposition data from tropical
37 forests (Jia et al. 2016; Vet et al. 2014) and the projected rise in atmospheric deposition of
38 nutrients, such as N and phosphorus (P) mainly due to anthropogenic activities, in tropical
39 regions highlights the need for empirical studies on this topic (Galloway et al. 2004; Wang et
40 al. 2017).

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

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

65 such as particles and adsorbed gases, and (III) bi-directional solute exchange between
66 intercepted rainfall and canopy surfaces like foliage, woody parts, epiphytes and
67 microorganisms (canopy exchange). Net throughfall is thus affected by bulk precipitation, dry
68 deposition and canopy exchange. Separating the contribution of dry deposition from canopy
69 exchange in throughfall has been an ongoing concern for several decades (Staelens et al.
70 2008), but is commonly calculated via the canopy budget model (Bredemeier 1988; Draaijers
71 et al. 1996; Ulrich 1983). This model distinguishes between internal and external input sources
72 to ecosystems using the so-called 'filtering approach' based on a tracer ion (Ulrich 1983) and
73 by estimating ion exchange processes occurring within the forest canopy (Draaijers et al.
74 1996).

75 Although the past two decades have seen an increase in research linking patterns of rainfall
76 and plant productivity in lowland tropical forests (Taylor et al. 2017) there is still a lack of
77 studies on atmospheric deposition in areas of high precipitation and temperature (Jia et al.
78 2016). Throughfall measurements from South American tropical forests are especially rare
79 (Bauters et al. 2019) and Vet et al. (2014) revealed that long-term measurements are even
80 rarer, with only a handful of studies reporting rainfall chemistry for periods of one year or
81 longer in the neotropics (excluding Central America).

82 Here, we present the results of a study spanning 15 months and report on annual fluxes of
83 bulk deposition and throughfall in a tropical forest in French Guiana. We compared the input
84 of external nutrients by bulk deposition to the internal recycling by litterfall and throughfall.
85 For elements such as phosphorus (P) and potassium (K), that have often been reported to be
86 limiting in tropical rain forests (Sardans and Peñuelas 2015; Turner et al. 2018; Wright et al.
87 2011), we expected a relatively large amount of bulk deposition compared to the litterfall and

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

91 **Materials and Methods**

92 **Site description**

93 The study was conducted at the Paracou research station (5°15'N, 52°55'W), 15 km from the
94 coast and about 40 km West from the nearest city, Kourou, in French Guiana. Annual rainfall
95 (2004 - 2015) averaged 3100 ± 70 mm yr⁻¹ and annual air temperature 25.7 ± 0.1 °C (Aguilos
96 et al. 2019). The tropical wet climate is highly seasonal due to the north/south movement of
97 the Inter-Tropical Convergence Zone, which brings heavy rains from December to July and a
98 very dry period from August to November (less than 100 mm precipitation per month). The
99 vegetation is highly diverse, with between 150 and 200 species ha⁻¹ for trees with diameter at
100 breast height > 10 cm. The most represented plant families occurring at the study site are the
101 Lecythidaceae, Fabaceae, Sapotaceae and Chrysobalanaceae (Gourlet-Fleury et al. 2004). Soils
102 in Paracou are mostly Acrisols (FAO 1998) developed over a Precambrian metamorphic
103 formation called the Bonidoro series. It is characterized by schists and sandstones and locally
104 crossed by veins of pegmatite, aplite and quartz (Epron et al. 2006). The landscape is
105 undulating with maximum slopes of approximately 15°. The elevation difference between hill
106 summits and valleys is 20 - 50 m over horizontal distances of 200 - 400 m.

107 **Experimental setup**

108 **Bulk and Throughfall precipitation**

109 Precipitation collectors were made of polyvinyl chloride (PVC) funnels (202 mm diameter, area
110 = 320.47 cm²) attached to an opaque PVC plastic tube (diameter 15 mm) inserted into a 5 L
111 polyethylene (PE) plastic bottle. Bottles were covered with aluminium foil, inserted into a 0.5

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

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

132 Littertraps

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

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

146 Chemical analyses

147 Chemical analyses were performed at the Ecosystem Management Research Group,
148 University of Antwerp, Belgium. Samples of the bulk precipitation and throughfall were
149 defrosted and an aliquot was filtered (0.20 µm) preceding the analysis of inorganic anions (Cl⁻
150 , NO₃⁻, SO₄²⁻, HPO₄²⁻) and cations (Na⁺, NH₄⁺, K⁺, Ca²⁺, Mg²⁺) by ion chromatography on an 883
151 Basic IC Plus (MetrOhm, Switzerland) with 919 IC Autosampler Plus (MetrOhm, Switzerland).
152 Nitrite (NO₂⁻) was also measured in these samples, but concentrations were consistently
153 below 1 µg L⁻¹ and are not discussed further. Anions were separated on a Metrosep A Supp 5
154 - 150 / 4.0 column with a Metrosep A Supp 5 guard / 4.0 protecting the separation column.
155 Cations were separated on a Metrosep C 6 - 150 / 4.0 column with a Metrosep C 4 guard / 4.0
156 protecting the separation column. All columns were produced and distributed by MetrOhm,
157 Switzerland. Data analysis was conducted with MagicIC Net software. For the determination
158 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).

180 **Definitions and calculations**

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

184
$$\text{VWM} = \frac{\sum(C_i * V_i)}{\sum(V_i)} \quad (1)$$

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

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

192
$$\text{ANF} = \sum_j^n \text{mean}(C_i * V_i)_j \quad (2)$$

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

196
$$TF = BP + CE + DD \quad (3)$$

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

199
$$\text{NTF} = TF - BP = CE + DD \quad (4)$$

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

202 Canopy Budget Model

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

$$218 \quad DD_X = \frac{(TF_{Na} - BP_{Na})}{BP_{Na}} * BP_X \quad (5)$$

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

220 Sea salt fraction

221 A common method used to quantify the inputs of marine chloride (Cl⁻), sulfate (SO₄²⁻)
222 magnesium (Mg²⁺), calcium (Ca²⁺) and potassium (K⁺) in rainwater is to compare the ionic

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

$$227 \quad [X]_{\text{nss}} = [X]_{\text{rain}} - \left([\text{Na}^+]_{\text{rain}} \times \left[\frac{X}{\text{Na}^+} \right]_{\text{SW}} \right) \quad (8)$$

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

233 **Statistical analysis**

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

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

255 Results

256 Bulk precipitation

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

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

267 There was variation in bulk precipitation ion concentration between the three collector
268 setups, with average CVs (N = 3) ranging between 14 and 40% for most ions, except for NH₄⁺

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

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

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

286 The percentage of each ion that was derived from sea salt (ss fraction) showed large variation
287 (Fig 2). Because we assumed it to be completely derived from sea salt (Berner and Berner
288 2012), the ss fraction of Na^+ was 100%. Surprisingly, the ss fraction of Cl^- was less than 50%,
289 indicating a large non-marine input. The ss fraction of Mg^{2+} exceeded 80%, while the ss
290 fractions of SO_4^{2-} , K^+ and Ca^{2+} were below 25%. We did not calculate the ss fraction of the N

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

293 Throughfall and canopy exchange

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

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

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

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

316 Litterfall

317 We captured around 30 kg of litter on a combined littertrap surface of $30.24 \pm 0.01 \text{ m}^2$
318 between May 17th 2016 and October fourth 2017. This translated into the production of $9.8 \pm$
319 0.2 Mg of leaf litter per hectare of forest over the study period, or $6.5 \pm 0.2 \text{ Mg ha}^{-1} \text{ y}^{-1}$ during
320 one year (from September 23rd 2016 until September 20th 2017). Litterfall showed large spatial
321 variation even within blocks, leading to within-block CVs ranging between 17 and 139%, with
322 an average CV of 55 and a median of 52%. Litterfall was greater during the months of June,
323 July, August, September and October compared with the rest of the year (Fig 4). Overall,
324 litterfall was highest in August ($0.75 \text{ Mg ha month}^{-1}$) and lowest in November (0.42 Mg ha
325 month^{-1}).

326 The carbon (C) content of the intercepted litter was, on average, $50.8 \pm 0.1\%$ over the entire
327 period. The concentrations of N and P in the leaf litter averaged at $12.1 \pm 0.1 \text{ g kg}^{-1}$ and 0.224
328 $\pm 0.008 \text{ g kg}^{-1}$. The average weight-based and molar C:N ratios were 42.0 ± 0.4 and 49.0 ± 0.5 ,
329 respectively, while the average weight-based and molar N:P ratios were 54 ± 2 and 119 ± 5 ,
330 respectively. The concentrations of K, Ca and Mg fell between the N and P concentrations,
331 with $1.6 \pm 0.1 \text{ g kg}^{-1}$, $6.42 \pm 0.07 \text{ g kg}^{-1}$ and $2.44 \pm 0.04 \text{ g kg}^{-1}$, respectively (Table 3). With the
332 exception of K, which was roughly equal in bulk precipitation and three times higher in
333 throughfall, the yearly deposition of each of the above measured macro-nutrients through
334 litterfall was greater than its deposition by rain or throughfall. Compared to litterfall, the bulk
335 precipitation and dry deposition together represented a large input of nutrients to the forest,
336 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

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

363 Nitrogen

364 The amount of TDN deposited by bulk precipitation was within the range reported for
365 primary tropical forests situated in Central and South America (Boy et al. 2008; Hofhansl et al.
366 2011; Wilcke et al. 2013). Bulk precipitation and dry deposition flux of N_{in} amounted to $\pm 2 \text{ kg}$
367 $\text{ha}^{-1} \text{ y}^{-1}$, placing our study site into the lower range of N_{in} deposition values reported for
368 lowland tropical forests, including a remote Central Amazonian site ($\sim 3 \text{ kg } N_{in} \text{ ha}^{-1} \text{ y}^{-1}$)
369 (Pauliquevis et al. 2012), and within the range of $1 - 2 \text{ kg } \text{ha}^{-1} \text{ y}^{-1}$ reactive N deposition
370 modelled by Wang et al. (2017) for 1997 until 2013. Anthropogenic sources of reactive N, such
371 as fossil fuel burning, livestock operations and agriculture (Dämmgen and Erisman 2005), are
372 scarce in French Guiana (Prospero et al. 2014), which may explain the low observed rates of
373 N_{in} deposition compared to other tropical sites in Central America (Hofhansl et al. 2011), Africa
374 (Galy-Lacaux and Delon 2014) or even Central Amazon (Andreae et al. 1990; Williams et al.
375 1997). There, often substantially higher rates of reactive N deposition were measured in
376 tropical sites situated within 20 to 100 km of a major city or industry.

377 In contrast to our low N_{in} , the amount of DON in bulk precipitation fell at the high end of
378 existing values reported for several South and Central American tropical forests (Boy et al.
379 2008; Hofhansl et al. 2011). However, DON values are not routinely measured and thus rarely
380 reported, resulting in a specific knowledge gap for tropical forests (Jia et al. 2016). A possible
381 source of DON at our site could be Amazon biomass burning, but the prevailing north-east
382 wind direction in French Guiana (Aguilos et al. 2019) renders a substantial contribution of this
383 N source unlikely. Instead, it is possible that at least a part of the deposited DON originated in

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

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

404 Phosphorus

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

408 deposited TDP was P_{in} , which is much higher than the global average 40% P_{in} contribution to
409 TDP (Tipping et al. 2014). This suggests that the contribution of P derived from biogenic
410 particles (including pollen, spores, bacteria, fungi, and fragments of leaves, which contain P in
411 predominantly organic form) was generally lower than the global average. Instead, this ratio
412 is indicative of dust deposition as soluble P in dust is primarily found in inorganic form (Graham
413 et al. 2003). Typically, soil dust particles originate from local sources within the forest, but
414 moist soils just like the soils at our site are less susceptible to wind erosion. Another likely
415 source of P containing dust are the trade winds, which carry soil dust mostly originating from
416 the Bodélé Depression in northern Chad (Ben-Ami et al. 2010; Koren et al. 2006; Swap et al.
417 1992). This dust contains, among other elements, P (Zamora et al. 2013) and has been
418 proposed as a major nutrient input into the Amazon (Yu et al. 2015). A recent study calculated
419 that, on average, both African biomass burning and dust transported across the Atlantic Ocean
420 provides $80 \text{ g P ha}^{-1} \text{ y}^{-1}$ into the Amazon (Barkley et al. 2019), but this value is averaged for the
421 entire Amazon basin and is likely higher in French Guiana than deeper in the continent
422 (Prospero et al. 2014; Yu et al. 2015). A recent study in Panama even found that in periods
423 with highest Saharan dust deposition up to $88 \pm 31 \text{ g P ha}^{-1} \text{ month}^{-1}$ arrives from Africa (Gross
424 et al. 2016).

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

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

448 On the ecosystem scale, the P input to the forest floor by litterfall is low at only 1.5 kg ha⁻¹
449 y^{-1} and, compared to the input of external P by atmospheric deposition, is equivalent to
450 roughly one third of the litterfall P (Figure 5). Relatively, the external input of atmospheric P
451 compared to the internal recycling by litterfall P was similar to what was found in Ecuador
452 (Wilcke et al. 2019), but higher than in other tropical forests in Panama (Gross et al. 2016) or
453 Costa Rica (Hofhansl et al. 2011). In Panama Saharan dust deposition atmospheric P input was
454 equivalent to between 10 and 29% of the P in monthly litterfall while in Costa Rica atmospheric
455 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.

479 The large amount of leached K^+ (Table 3) is in line with what is found in other forests
480 around the world where K^+ deposition and subsequent movement through the canopy
481 generally results in the net removal of K from canopy surfaces in all biomes (Ponette-González
482 et al. 2016). Not only is dry-deposited K^+ highly soluble in water, but internal plant K^+ is
483 concentrated in cells near leaf surfaces (Schlesinger and Bernhardt 2013) and thus sensitive
484 to leakage following, e.g., cuticular damage. As a result, K^+ is easily washed from canopy
485 surfaces and leached from plant tissues; thus K^+ leaching is generally high in wet environments
486 such as tropical forests, sometimes reaching up to $147 \text{ kg } K^+ \text{ leached ha}^{-1} \text{ y}^{-1}$ (Boy and Wilcke
487 2008).

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

494 Chloride

495 Both Cl^- and Na^+ are the most abundant ions in seawater, between them constituting \pm
496 85% of the total salinity in the oceans, and it is commonly accepted that the primary source
497 of atmospheric chloride comes from the ocean. In areas close to the sea the Na^+/Cl^- ratio in
498 precipitation is typically that of sea salt (Keene et al. 1986), thus the sea salt fraction of Cl^- in
499 these coastal areas is typically close to 100%. This was not observed at our site where Cl^-
500 deposition fluxes far exceeded those of Na^+ and the sea salt fraction of Cl^- was just below 50%,
501 which is highly surprising for a forest situated close to the Ocean. French Guiana has few of
502 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).

549 Conclusion

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

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

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

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

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580 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

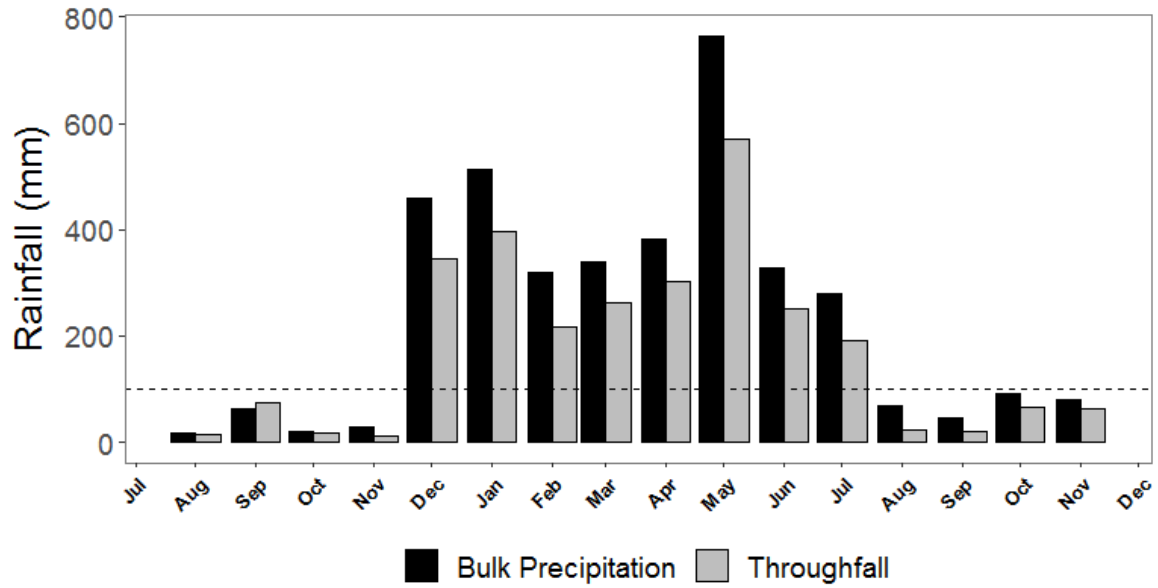


Figure 1 Mean monthly precipitation measured in the bulk precipitation (black) and throughfall (grey) over the period August 19th 2016 until November 15th 2017. The horizontal dashed line indicates a precipitation of 100 mm and all months with lower precipitation are considered dry season months

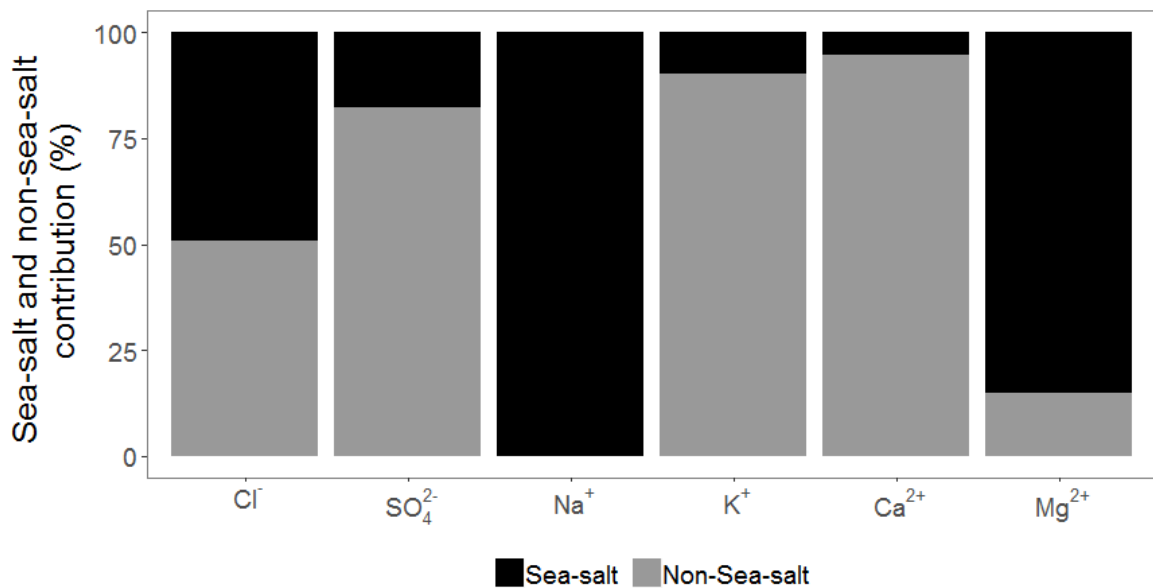


Figure 2 The marine (sea-salt, black) or non-marine (non-sea-salt, grey) origin of Cl⁻, SO₄²⁻, K⁺, Ca²⁺ and Mg²⁺ expressed as a percentage, assuming all Na⁺ was marine in origin (see Berner and Berner 2012).

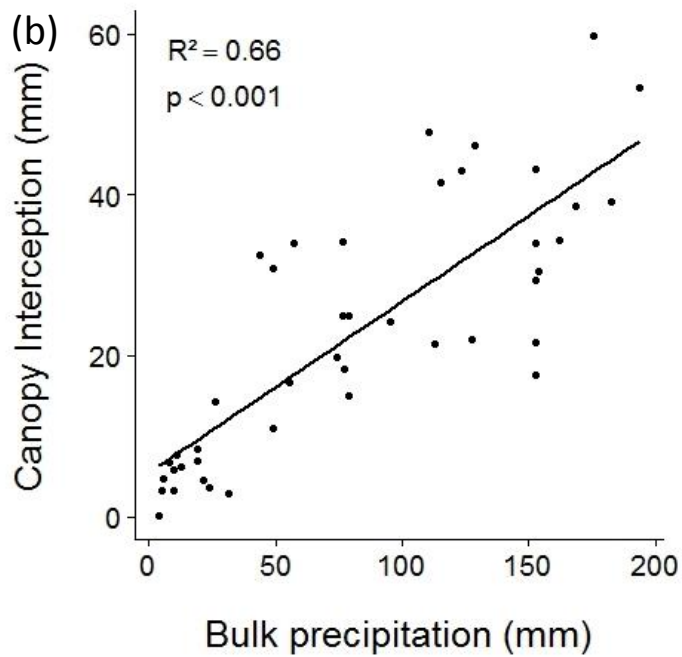
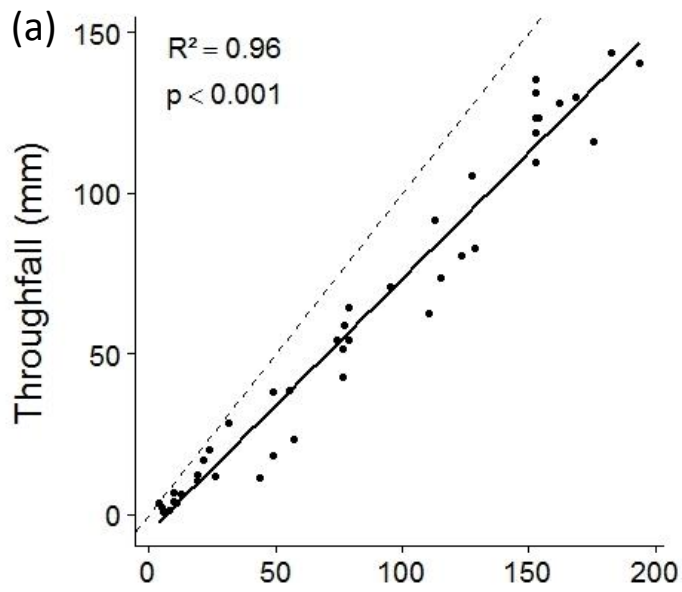


Figure 3 Relationship between (a) bulk precipitation and the throughfall volume measured for a given sampling period, and (b) bulk precipitation and the precipitation volume intercepted by the canopy. The 1:1 line in graph (a) is given by the dashed black line. Both the R^2 and the significance of the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

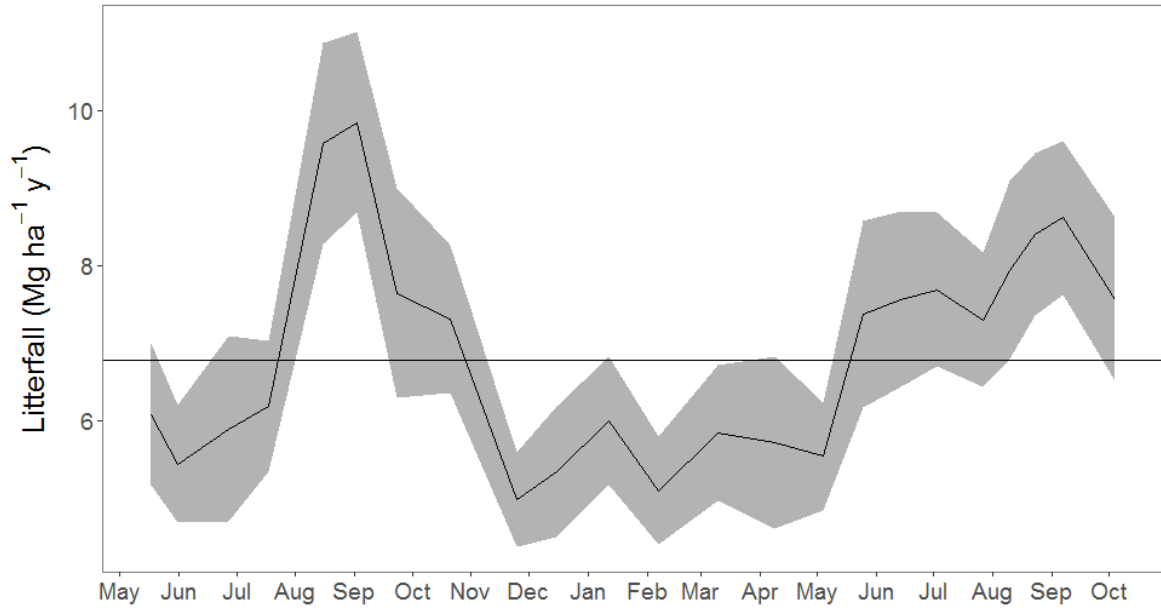


Figure 4 Litterfall over the entire study period. The 95% confidence interval is depicted by the grey band and the overall average litterfall is depicted by the horizontal line. Note that the y-axis does not start on 0.

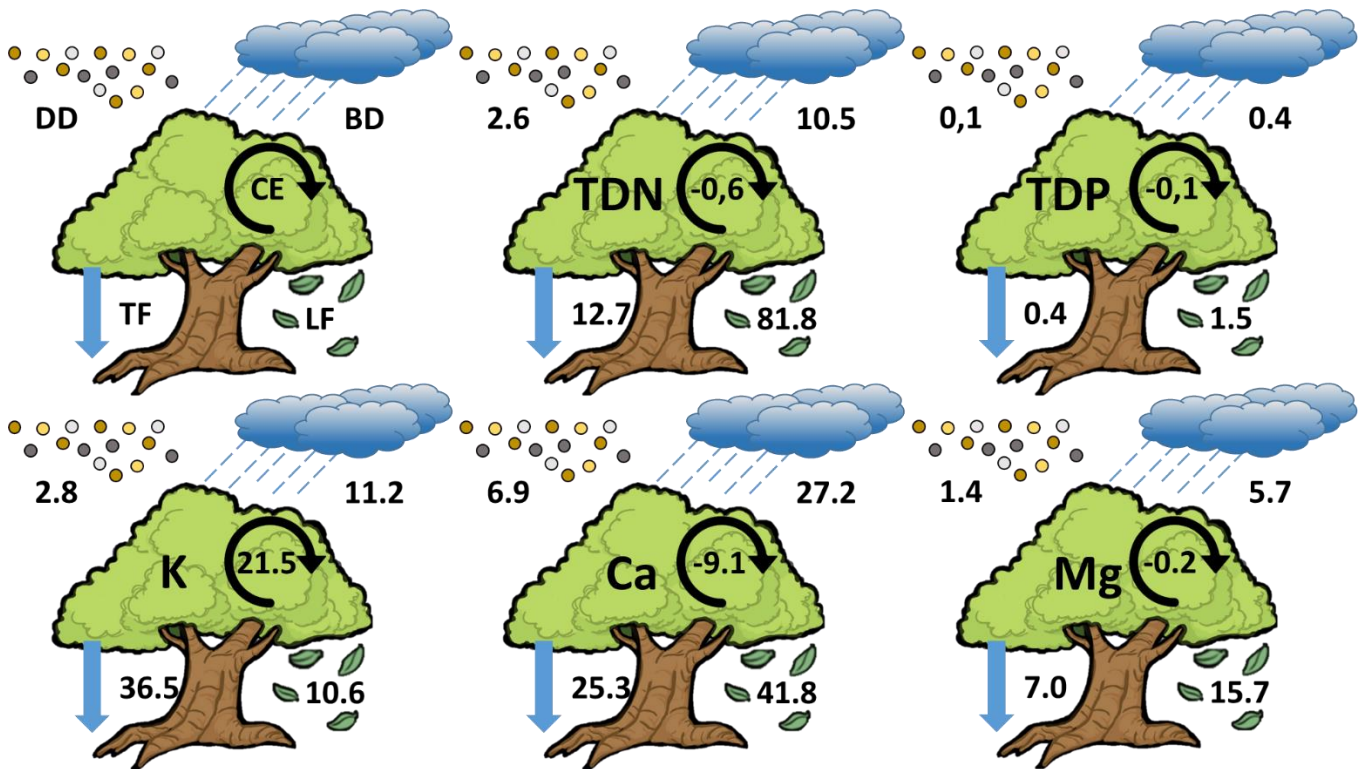


Figure 5 Overview of the inputs, outputs and canopy exchange measured in this study for total dissolved nitrogen (TDN), total dissolved phosphorus (TDP), potassium (K), calcium (Ca) and magnesium (Mg). Negative values for CE indicate canopy uptake and positive values indicate canopy leaching. Value units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

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