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Assessment of Wet Inorganic Nitrogen Deposition in an Oil Palm Plantation-Forest Matrix Environment in Borneo

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Abstract: Nitrogen (N) deposition significantly affects forest dynamics, carbon stocks and biodiversity, and numerous assessments of N fluxes and impacts exist in temperate latitudes. In tropical latitudes, however, there are few such assessments. In this study, we measured the inorganic N concentration (wet deposition) deposited in rainfall and rainfall pH throughout one year at the boundary of a forest reserve in Malaysian Borneo. We considered that the N deposition may be either from forest and agricultural fires or derived from agricultural fertiliser. Therefore, we determined the wind trajectories using the HYSPLIT model provided by NOAA, the location of fires throughout the landscape throughout one year using NASA's FIRM system, and obtained the land use cover map of Malaysia and Indonesia. We then correlated our monthly cumulative wet N deposition with the cumulative number of fires and the cumulative area of oil palm plantation that wind trajectories arriving at our study site passed over before reaching the rainfall sampling site. At 7.45 kg N ha^{-1} year^{-1}, our study site had the highest annual wet inorganic N deposition recorded for a Malaysian forest environment. The fire season and the cumulative agricultural area crossed by the winds had no significant effect on N deposition, rainfall N concentration, or rainfall pH. We suggest that future research should use ^{15}N isotopes in rainfall to provide further information on the sources of N deposition in tropical forests such as this.

Keywords: Borneo; forest fires; haze; heath forest; nitrogen deposition; oil palm fertilisation

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1. Introduction

Forests in the tropics are composed of a matrix of different forests types. Whilst the widespread lowland evergreen rain forests are thought to be phosphorus (P) limited [1], other tropical forests such as heath and montane, are considered nitrogen (N) poor [2–4]. So, N poor forests may respond differently to increases in N inputs than lowland evergreen rain forests. The use of fertilisers, fossil fuel and biomass burning have been increasing the concentration of atmospheric reactive N (defined as oxide gases, anions and amine derivatives) since the mid-20th century in the industrialised northern hemisphere, with a consequent increase in N deposition [5,6]. The global dataset of [7] allows a comparison of worldwide N deposition rates ranging from 0.17 kg N ha^{-1} year^{-1} in Alaska to 27.1 kg N ha^{-1} year^{-1} in parts of China. Globally, more than 11% of non-agricultural vegetated land received an amount of wet and dry inorganic N deposition exceeding a critical load of 10 kg N ha^{-1} year^{-1} [8], above which a negative effect on vegetation is expected. Critical loads are set according to abiotic and biotic conditions affecting sensitivity. Acidic European coniferous forests, for example, are highly sensitive to N deposition and have a N critical load of 10–15 kg N ha^{-1} year^{-1} [9], whereas Chinese subtropical evergreen forests have a higher N critical load of 20–70 kg N ha^{-1} year^{-1} [10] because they are P limited. However, in the tropics, N deposition leads to a drop in soil pH [11] and decreases in plant biodiversity [12,13]. The rate of N deposition is predicted to increase in tropical areas [14]. The Sundaland biogeographic region that includes the island of Borneo, for example, is predicted to exceed a deposition rate of 10 kg N ha^{-1} $year^{-1}$ on 15.6% of its area by 2050 [14]. Despite the importance of tropical regions as biodiversity hotspots and the potentially damaging effects of N deposition, few monitoring stations for N deposition and studies on N deposition exist in these zones [7].

South-east Asia seasonally undergoes a burning season, usually around September-October [15], where land is cleared and burned to establish small-scale agriculture or agro-industrial plantations [16]. The greatest fire density has been recorded in Kalimantan and Sumatra, Indonesia [17] and, to a lesser extent, in Malaysian Borneo and Brunei [18]. Biomass burning in tropical regions produces a haze that is an important input source of atmospheric N [19–21]. Another potential source of reactive N to forests is agriculture. Agro-industrial crops such as oil palm [22] drive the demand of tropical countries for N fertilisers that consume c. 70% of the global fertiliser production [23]. Few measurements of N deposition exist for Malaysian Borneo (but see [7]), where rain forests are enclosed in a mosaic of oil palm estates [24] and are likely to undergo seasonal haze pollution from fires and N deposition due to the fertilisation of plantations. Borneo has widespread N-limited forests such as heath forests (known as kerangas) and montane forests. Both of these forest types host a high plant endemism with species adapted to low soil nutrients [25]. These forests are, therefore, important to maintain the biodiversity of tropical areas overall and of the island of Borneo in particular. An increase in N deposition could thus lead to biodiversity losses in a similar way to temperate ecosystems. In this research, we hypothesised that, as our study site is near to an intensive agricultural area and receives wind from areas that suffer fires, it is subject to a high amount of N deposition. We tested this by collecting rainfall with a wet-only sampler throughout one year and measuring the concentrations of rainfall NH_4^+ and NO_3^-. Furthermore, we hypothesised that the monthly amount of wet N deposited at our study site from rainfall is correlated with fire occurrences or N fertilizer addition to nearby cultivated land that is brought to the site by the prevailing winds. We tested this second hypothesis by back-crossing the monthly wind trajectories arriving at our study site with the monthly fires map obtained by satellite and the land use map of the neighbouring area. In this way we obtained two indices that were then correlated with the monthly deposition of wet inorganic N to determine their relationship.

2. Materials and Methods

2.1. Study Area

Our collection site was located at the Sabah Forestry Department's Forest Research Centre (FRC) in Sepilok, Sabah, Malaysia (Figure 1). The FRC building is surrounded by a matrix of residential dwellings, fruit orchards and oil palm estates, and there is an urban centre (Sandakan) about 10 km to the east. Further south from FRC (c. 1 km) and west (c. 0.5 km) extends a 44 km^2 primary forest growing on alluvial, sandstone and heath forest soil [26], the Kabili-Sepilok Forest Reserve. The close proximity of the forest reserve to the matrix of urbanized and agricultural landscape is typical for northern Borneo, given the rapid expansion of agroindustry and human activities. The climate is equatorial with a mean temperature of c. 26 °C and an annual rainfall of c. 3000 mm [27,28]. Most of the rain falls between November and February due to the influence of the north-east monsoon, which brings winds from the Philippines. The driest month is usually April, when the rainfall is less than 100 mm $month^{-1}$ [28]. From June to August, the south-west monsoon brings air masses from Indonesian Kalimantan, southern Borneo [27].

2.2. Sampling and Analysis

Nitrogen deposition comprises wet and dry depositions; in this study, we were only able to sample wet N deposition. We collected rainwater with a wet-only sampler placed on the highest point of the FRC building. The sampler was composed of two cylinders one inside the other; the two cylinders were connected by a funnel. This allowed even the lightest rain to be collected in the sampler and reduced the evaporation. Sampling was carried out twice per week over a period of 12 months from September 2016 to August 2017. We recorded rainwater volume using a rain gauge. Sub-samples of rainfall were then filtered with a 0.2 μm filter. After filtration, we recorded rainwater pH with a Corning 240 pH meter, then we acidified the samples to pH ~2 with two drops of 50% H_2SO_4. Samples were then stored in a −20 °C refrigerator until NH_4^+ and NO_3^- analyses could be carried out using a segmented flow analyser (Astoria-Pacific A2, Clackamas, OR, USA). We did not measure total dissolved N, so our results refer to wet-deposited inorganic N only. We calculated monthly deposition of N derived from NH_4^+ and NO_3^- (referred as NH_4^+-N and NO_3^--N) as well as the monthly volume-weighted concentration of NH_4^+ and NO_3^-. Volume-weighted concentration is a routine measure that expresses the average concentration of ions in the total volume of precipitation that fell within a certain time period. Volume weighted concentration (Ci) was calculated as

$$Ci = \Sigma jPjCij/\Sigma jPj$$

where Pj is the amount of precipitation in sampling j (mm) and Cij is the concentration of constituent i for sampling j (μmol l^{-1}) [29]. We then multiplied the volume-weighted N concentration by the total precipitation for each month to obtain the bulk wet deposition in kg ha^{-1} $year^{-1}$.

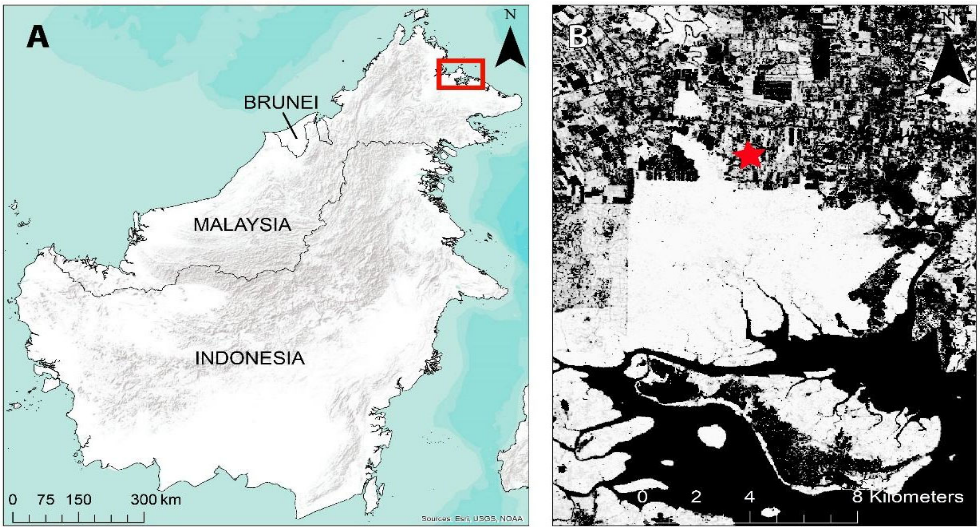

Figure 1. Location of the study site. (**A**) The island of Borneo with the approximate position of the Kabili-Sepilok Forest Reserve (Sabah, Malaysia) highlighted in red. (**B**) The Kabili-Sepilok Forest Reserve with the location of the Forest Research Centre (red star) highlighted. Panel B shows forested areas in white and non-forest areas in black (data from [30]).

2.3. Wind Trajectories, Fires, Land Use and the Correlation with N Deposition

We considered fires and agricultural activities as providing atmospheric N inputs and winds as N transporters. To investigate whether active fires or land use were a significant source of wet N deposition, we back-crossed wind trajectories with fire maps for the study area generated using the HYSPLIT model provided by NOAA (www.ready.noaa.gov, accessed on 20 October 2022; [31]) and with the land use map of year 2016 from the Nusantara Atlas (https://map.nusantara-atlas.org/ accessed on 19 December 2019; [32]) using ESRI ArcMap (version 10.3.1). To this aim we used the Quantun Geographic Information System QGIS [33]. Throughout the study period (September 2016 to August 2017), we generated twelve wind trajectories (width of 12 km) every two weeks at three different altitudes (50 m, 500 m and 1000 m), each running for 315 h. We chose these wind heights because the smoke released from fires in Borneo has been estimated to reach heights between 523 m and 993 m [34]. Nonetheless, smoke is concentrated at a lower height so we also added the 50 m trajectory. We obtained the fire maps from NASA's FIRMS system, which provides data from active fires detected by MODIS C6 satellites (https://earthdata.nasa.gov/earth-observation-data/near-real-time/firms accessed on 3 December 2018). Using these data, we counted how many fires the winds arriving at our study site passed over every two weeks. We used only fire points which had a detection confidence over 30%, as suggested by the MODIS website (https://www.earthdata.nasa.gov/faq/firms-faq#ed-confidence accessed on 22 December 2019). The smoke from each fire becomes less dense with increasing distance from the fire itself. This is of course a simplification and it depends on the meteorology at the time of the fire and on the air mass movements. We accounted for this by creating three buffer rings around each fire point. The rings had a radius of 5, 10 and 15 km from the fire point and we assigned a lower weighting to the largest 15 km ring, an intermediate weighting to the 10 km radius ring and the greatest weighting to the 5 km ring. We then used the 'intersect' command in QGIS to assign to each wind point the weight of the respective fire buffer ring that it passed over. To assess the type of land use that the winds arriving to our sampling point were passing over, we used the 'join attributes by location' function in QGIS. Among the wind points passing over oil palm plantations, we assigned a value of 1 to the wind points at 50 m height, a value of 0.6 to the wind points at 500 m height and a value of 0.3 to the wind points at 1000 m height. Finally, we calculated the cumulative monthly fire count (MFC) and the cumulative monthly land use count (MLUC) to assess if there was a relationship between changes in monthly total wet N deposition and concentration in rainfall with the potential air pollution derived from fires or from agricultural fertilisers. Given the normal distribution of our variables (Shapiro–Wilk's test), we examined the correlations of MFC and MLUC with monthly rainfall, rainwater pH, NH_4^+-N and NO_3^--N total monthly deposition, and NH_4^+ and NO_3^- concentration in rainfall, with Pearson correlation tests. All the *p*-values were adjusted using the Benjamini–Hochberg false discovery rate (fdr) procedure [35]. We also conducted a sensitivity analysis to make sure that assigning different weights to the buffer rings and the winds did not alter the significance of the Pearson tests. All the statistical analyses were performed with R version 3.5.1 [36].

3. Results

Over the 12 months of monitoring (from July 2016 to August 2017), we recorded 4637 mm total rainfall, which was higher than the mean of around 3000 mm year^{-1} for this location ([27]; www.ncdc.noaa.gov/cdo-web/ accessed on 22 December 2019). During this time, the driest month was, unexpectedly, February, with 175 mm rainfall, and November was the wettest month (765 mm; Figure 2). The monthly mean volume-weighted concentration of NH_4^+ in rainfall was lowest in October 2016 (3.5 μmol L^{-1}) and peaked in June 2017 (21.6 μmol L^{-1}), declining again in the following two months (Table 1). June 2017 also had the greatest wet NH_4^+-N deposition (1.28 kg N ha^{-1}); the lowest was in October 2016 (0.21 kg N ha^{-1}). The volume-weighted concentration of NO_3^- remained constant through the year (1.1 μmol l^{-1} ± 0.09 SE; Figure 2). In total, we calculated a deposition rate of wet inorganic N of 7.45 kg N ha^{-1} year^{-1} which was dominated by NH_4^+ (90.4%), with NO_3^- making up the remainder (9.6%). Rainfall had an annual mean pH of 5.38 ± 0.02 SE, with the most acidic rain event in October 2016 (pH 4.71) and the least acidic in August 2017 (pH 5.82).

The winds coming from the Philippines are characteristic of the north-east monsoon, usually from November to February, although these winds were still active until April 2017 (Figure 3). The south-west monsoon usually appears around June and July but, according to our simulation, these months had quite weak winds; strong south-west monsoon winds arrived only in August. Throughout our study period, two major fire seasons occurred in the region. The first fire season was between September and October 2016 with high concentrations of fires in Kalimantan and Sarawak. The second was between March and

May 2017, with fires in the western part of the Philippines (i.e., Palawan, Western Visayas and Negros island, as well as Zamboanga peninsula; Figure 3).

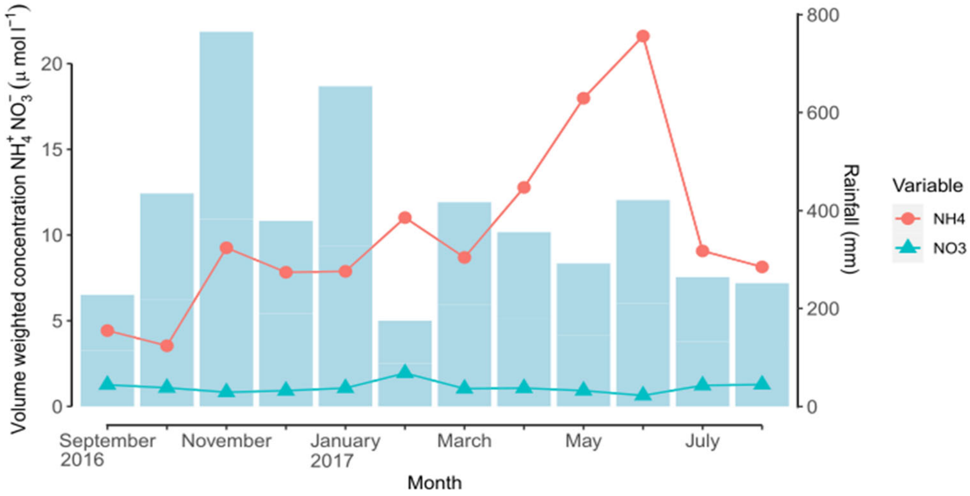

Figure 2. Total monthly rainfall at our study site in Sepilok, Sabah, Malaysia (blue bars) and volume-weighted mean concentration of rainfall NH_4^+ and NO_3^-.

Table 1. Monthly mean pH and the cumulative amount of wet NH_4^+-N and NO_3^--N (kg ha^{-1}) in rainfall collected in our study site at Sepilok, Sabah, Malaysia.

Date		pH	NH_4^+-N	NO_3^--N
2016	September	5.37	0.35	0.08
	October	5.17	0.21	0.07
	November	5.38	0.99	0.09
	December	5.30	0.41	0.05
2017	January	5.44	0.72	0.09
	February	5.45	0.27	0.05
	March	5.34	0.51	0.06
	April	5.44	0.64	0.05
	May	5.46	0.73	0.04
	June	5.39	1.28	0.04
	July	5.32	0.33	0.04
	August	5.47	0.29	0.04
Total		-	6.74	0.71

The winds arriving at our study site in September 2016 passed over the greatest number and intensity of fires; the winds arriving in December 2016 passed over the least number and intensity of fires. Winds from September and October 2016 arriving at our site intercepted fires from both Sarawak (at approximately 500 km distance) and Kalimantan (at approximately 450 km distance), whereas during the last two months of the north-east monsoon (March and April), winds intercepted the fires in the south-west Philippines, which had been detected by the satellites over a period of approximately nine months.

The winds that arrived at our study site in August 2017 passed through the greatest amount of oil palm cultivated areas (MLUC of 1492), whereas the winds arriving at our study site in February 2017 passed through the smallest extent of oil palm cultivated areas (MLUC of 101). We found no significant correlation between any N form or rainfall pH with MFC or and the MLUC, either with and without applying the fdr correction (Table 2; $p > 0.05$ in all cases).

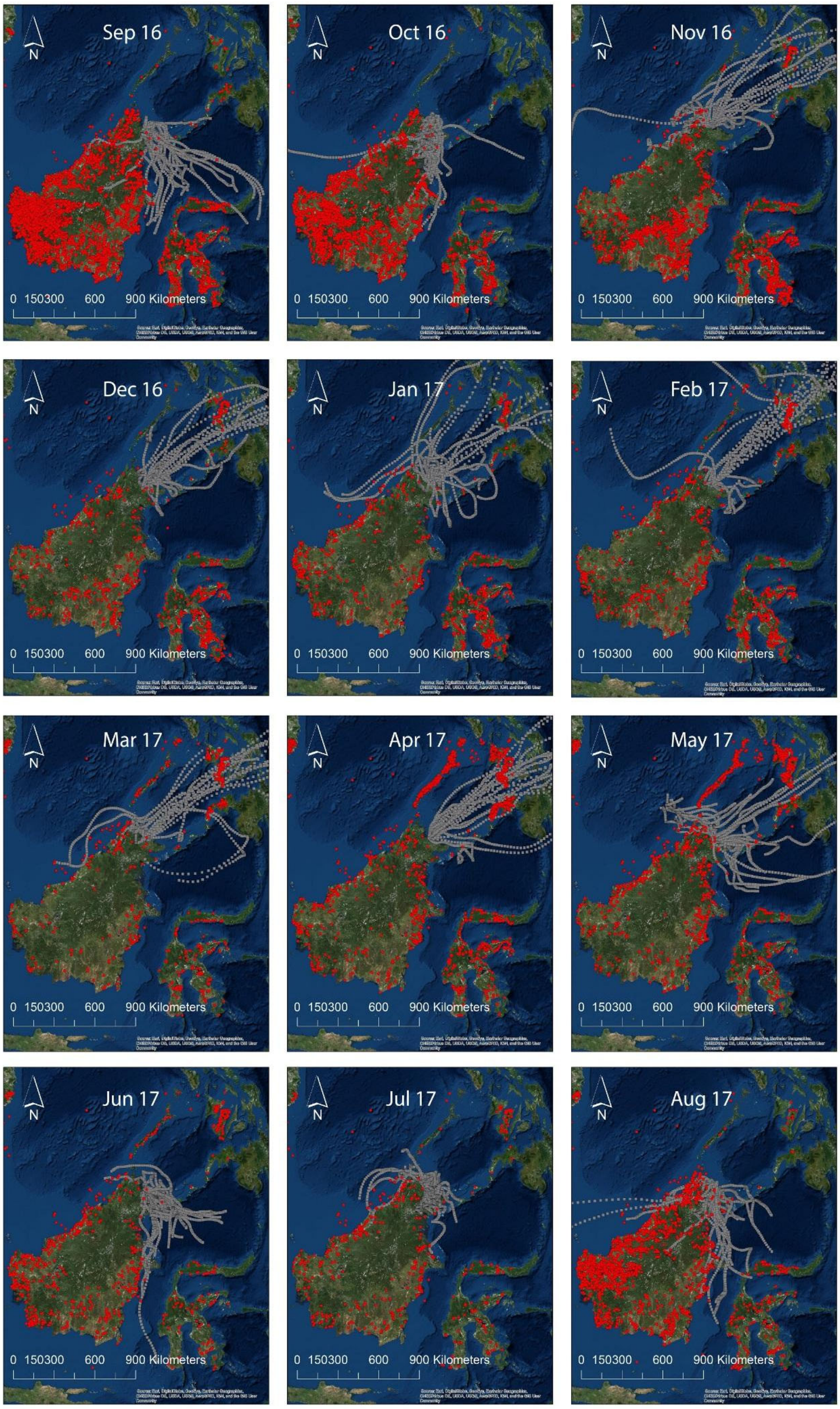

Figure 3. Intersection of the fire map (where the fires are represented by the red dots) obtained for Borneo and surrounding regions (MODIS C6 satellites https://earthdata.nasa.gov/earth-observation-data/near-real-time/firms accessed on 3 December 2018) with the backward wind trajectories (grey dotted lines) generated with the HYSPLIT model provided by NOAA. Backward wind trajectories were generated every two weeks at three different altitudes (50 m, 500 m and 1000 m) each running for 315 h. The backward wind trajectories end at our sample collection site in Sepilok, Sabah, Malaysia.

Table 2. Pearson correlation coefficients between the amount of fires that winds arriving at our sampling site passed over in the month before rainfall (MFC), the amount of oil palm cultivated area that winds arriving at our sampling site passed over in the month before rainfall (MLUC) and different monthly rainfall characteristics (total NH_4^+-N and NO_3^--N deposition, NH_4^+ and NO_3^- mean concentration in rainfall, mean rainwater pH and rainfall amount). Rainfall samples were collected throughout one year at the Forest Research Centre, Sepilok, Sabah, Malaysia. The *p*- values (adjusted using the Benjamini–Hochberg false discovery rate are noted in superscript and were all non-significant.

	NH_4^+ kg ha^{-1}	NO_3^- kg ha^{-1}	NH_4^+ µmol L^{-1}	NO_3^- µmol L^{-1}	Rainwater pH	Rainfall (mm)
MFC	$-0.30^{0.34}$	$0.27^{0.42}$	$-0.22^{0.48}$	$0.25^{0.38}$	$0.04^{0.90}$	$-0.37^{0.24}$
MLUC	$-0.04^{0.90}$	$0.21^{0.35}$	$-0.09^{0.78}$	$-0.30^{0.51}$	$-0.19^{0.55}$	$-0.31^{0.32}$

4. Discussion

Nitrogen deposition is known to decrease plant diversity in terrestrial N-limited ecosystems [37,38]. This evidence mainly comes from studies of temperate sites and tropical forests are underrepresented. Here, we assessed the amount of N deposition over one year for a single location in Sabah, Malaysian Borneo, where forests are in close proximity to N-emitting sources such as fires and agro-industrial cultivation. We also assessed the relationship of N deposition with the trajectory of winds passing over fires and oil palm cultivation, as both are known to emit N. At 7.45 N ha^{-1} year^{-1}, our study site received the highest wet inorganic N deposition measured for a forest environment in Malaysia (Table 3). Previous measurements have been carried out in lowland pristine forest (1.1 to 2.2 kg N ha^{-1} year^{-1}, [7,39]), forest surrounded by oil palm and rubber cultivation (the highest measurement of 6.2 kg N ha^{-1} year^{-1}, [40,41]) and montane forest (3.0 to 3.7 kg N ha^{-1} year^{-1} [7,42]). As we measured only wet inorganic N deposition and were unable to consider organic N and dry deposition (which can be equal to or exceed wet N deposition), it is likely that the total N deposition in our study site exceeds the critical load of 10–15 kg N ha^{-1} year^{-1}, that is proposed as potentially damaging to temperate forests [38]. This is relevant because heath forest, a forest typology widespread throughout the tropics, is possibly limited by soil N, similar to temperate forests [43,44]. Heath forest, thus, could have a critical load similar to temperate forests. Mean rainfall pH was mainly high and characteristic of an environment with low emissions of oxidised N, although pH sometimes fell below 5.3, indicating acidic deposition [7]. Given the low and stable oxidised N concentration recorded during our monitoring, this pH variation might be driven by fluctuations in sulphur concentration [45], which we did not measure. We acknowledge that the use of only one rain gauge collector and the limited sampling of only 12 months poses a limitation to the statistical power of our study. The rainfall pattern during the 12 months of sampling (from July 2016 to August 2017) was not typical for the year as it had c. 1600 mm of rain more than the average year. Nonetheless, our study site, being between a forest edge and a matrix of agro-industrial cultivation and human settlements, is representative of a large share of tropical rainforests, given that 20% of tropical forests lies within 100 m of a forest edge [46].

The pattern of NH_4^+-N concentration in rainfall suggested seasonal wet N deposition variation, but we were not able to pinpoint the origin of this seasonal variation. The seasonal pattern of fires we detected is typical for Borneo, with most active fires in September [47]. The large number of fires in south-west Philippines is concomitant with the dry season for the area (Philippine Atmospheric, Geophysical and Astronomical Services Administration, http://bagong.pagasa.dost.gov.ph/ accessed on 22 December 2019), suggesting a seasonal fire regime. However, despite the known emission of oxidised and reduced N from fires [48], we did not find any correlation between monthly wet inorganic N deposition or concentration in rainwater and the number of fires that winds coming to our study site intercepted. The winds from the month with the highest rainwater NH_4^+ concentration

(June 2017) intercepted the least number of fires. Biomass combustion generally produces oxidised N [19,49], which was low in our rainfall samples (9% of the total N deposition) and stable throughout the year. We thus consider it unlikely that fires and haze affected N deposition at our study site. We also suggest that the most reactive N deposited on these forests originates from nearby sources, a reasonable hypothesis in many tropical forests due to the heavy rainfall and high interception area by dense leafy canopies [50].

Table 3. Comparison of annual wet inorganic N deposition rates in some Malaysian and Singaporean sites.

Site		**NH_4^+-N**	**NO_3^--N**	**pH**	**Rainfall**	**Reference**
		kg ha^{-1} year^{-1}			**mm year^{-1}**	
Forest	Forest type					
Sepilok (East Malaysia)	Lowland	6.7	0.7	5.4	4637	This study
Danum (East Malaysia)	Lowland	0.7	0.4	5.1	2996	Vet et al., 2014 [7]
Pasoh (West Malaysia)	Lowland	2.3	3.9	5.8	2381	Manokaran 1980 [40]
Berembun (West Malaysia)	Montane	1.4	1.6	5.9	1979	Yusop and Nik 1989 [42]
Tanah Rata (West Malaysia)	Montane	1.4	2.1	4.9	2879	Vet et al., 2014 [7]
City						
Singapore		11.5	13.4	-	2554	Karthikeyan et al., 2009 [51]

We suggest that the main source of NH_4^+ in rainwater in these forests is agro-industrial fertilisation. Oil palms are regularly fertilised. We hypothesised that this fertilisation produces large amounts of ammonia (NH_3) and other reduced N compounds (collectively, NHx) that are hydrolysed to ammonium (NH_4^+) in the lower atmosphere and deposited in rainfall and dry deposition to downwind forests. Nonetheless, we found no correlation between any form of wet N deposition with the amount of oil palm plantations that the winds arriving at our study site intercepted. We speculate that the amount of N emitted by plantations, and thus the amount of N deposited by rain, is highest immediately after the fertilisation. It would then be necessary to know the seasonality of N fertilisation in oil palm plantations. There are, to our knowledge, no studies of NHx emission and deposition from oil palm plantations, although it is known that these compounds are, in general, produced in abundance after crop fertilisation [38,52]. In addition, one study found that nitrous oxide (N_2O) emissions, also presumably from fertilisation, can increase about 160 times from natural forests to oil palm plantations [53]. We hypothesise that the relatively still air throughout May and June could have led to an accumulation of fertiliser-derived NH_4^+ in the air mass that was then deposited in rainwater. In order to better understand the origins and the consequences of N deposition on Bornean forests, further work should investigate the seasonality of oil palm fertilisation.

5. Conclusions

Excess N deposition poses hazards to N-limited forest ecosystems such as the heath and montane forests in Borneo, potentially causing soil acidification, eutrophication, damage to roots, foliage, and mycorrhizas, species composition change, and biodiversity loss [38]. Here, we assessed the magnitude of wet inorganic N deposition over one year (2016–2017) at a site close to a pristine forest reserve. The site is also next to oil palm plantations and might be affected by volatilised N fertiliser from the plantation. It may also be impacted by the reactive N in seasonal haze from forest fires.

Our site received an amount of wet inorganic N deposition higher than any previously recorded from Malaysian forests. The deposition was mainly composed of NH_4^+ that increased substantially from April through June 2017. We did not find evidence that forest fires nor the passage of winds over oil palm plantations influenced the concentration or the total deposition of inorganic N. We consider that changes in rainwater NH_4^+ concentration were due to NH_4^+ accumulation sourced from the surrounding fertilised oil palm estates, rather than from forest fires. To further test this, we can compare the isotopic N signature of fertilisers used in the area to determine the source of N deposited on the forest. Furthermore, it should be clearly defined in which months the fertiliser is applied to the oil palm sites. Should we discover that fertiliser is the main contributor, then policy makers and local authorities should consider limiting the amount of fertiliser used in plantations near threatened forest sites.

Author Contributions: Conceptualisation, G.S., F.Q.B., J.T., N.B.D. and C.D.F.; methodology, G.S., N.M. and R.R.; formal analysis, G.S., S.E.P. and D.C.; resources, N.M. and R.R.; writing—original draft preparation, G.S., F.Q.B. and J.T.; writing—review and editing, All authors. All authors have read and agreed to the published version of the manuscript.

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