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LETTER

Biomass burning drives atmospheric nutrient redistribution within forested peatlands in Borneo

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Abstract

Biomass burning plays a critical role not only in atmospheric emissions, but also in the deposition and redistribution of biologically important nutrients within tropical landscapes. We quantified the influence of fire on biogeochemical fluxes of nitrogen (N), phosphorus (P), and sulfur (S) in a 12 ha forested peatland in West Kalimantan, Indonesia. Total (inorganic + organic) N, NO_3^-–N, NH_4^+–N, total P, PO_4^{3-}–P, and SO_4^{2-}–S fluxes were measured in throughfall and bulk rainfall weekly from July 2013 to September 2014. To identify fire events, we used concentrations of particulate matter (PM_{10}) and MODIS Active Fire Product counts within 20 and 100 km radius buffers surrounding the site. Dominant sources of throughfall nutrient deposition were explored using cluster and back-trajectory analysis. Our findings show that this Bornean peatland receives some of the highest P (7.9 kg PO_4^{3-}–P $ha^{-1}yr^{-1}$) and S (42 kg SO_4^{2-}–S $ha^{-1}yr^{-1}$) deposition reported globally, and that N deposition (8.7 kg inorganic N $ha^{-1}yr^{-1}$) exceeds critical load limits suggested for tropical forests. Six major dry periods and associated fire events occurred during the study. Seventy-eight percent of fires within 20 km and 40% within 100 km of the site were detected within oil palm plantation leases (industrial agriculture) on peatlands. These fires had a disproportionate impact on below-canopy nutrient fluxes. Post-fire throughfall events contributed >30% of the total inorganic N (NO_3^-–N + NH_4^+–N) and PO_4^{3-}–P flux to peatland soils during the study period. Our results indicate that biomass burning associated with agricultural peat fires is a major source of N, P, and S in throughfall and could rival industrial pollution as an input to these systems during major fire years. Given the sheer magnitude of fluxes reported here, fire-related redistribution of nutrients may have significant fertilizing or acidifying effects on a diversity of nutrient-limited ecosystems.

1. Introduction

Coupled changes in climate and land use are altering fire regimes in tropical forest regions (Siegert *et al* 2001, Cochrane 2003, Bowman *et al* 2009, Hansen *et al* 2009, Margono *et al* 2014), with consequences for global, regional, and local biogeochemical cycles (Crutzen and Andreae 1990, Andreae and Merlet 2001, van der Werf *et al* 2010). For example, over the past two decades, Indonesia has transitioned from a historically long fire-return interval regime (Goldammer 2007) to become one of the most important sources of biomass burning emissions worldwide (van der Werf *et al* 2010). In 2015, carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) CO_2-equivalent emissions from Indonesian fires were estimated to exceed Japan's 2013 fossil fuel CO_2 emissions (Global Fire Emissions Database 2015).

Although fires in Indonesia are anthropogenic—ignited during drier conditions primarily to clear vegetation for agriculture—these land-clearing fires and associated wildfires have increased dramatically in Sumatra and Kalimantan since the early 1990s as a direct result of governmental land-use policies, particularly logging and forest degradation, economic incentives for industrial agriculture, and government-sponsored transmigration, among other factors (Siegert *et al* 2001, Curran *et al* 2004, Dennis *et al* 2005, Langner *et al* 2007, Murdiyarso and Adiningsih 2007, Langner and Siegert 2009, Page *et al* 2009). El Niño Southern Oscillation (ENSO) conditions exacerbate dry season droughts and result in nonlinear increases in fire activity (van der Werf *et al* 2008). Mounting evidence suggests that fire regimes in Sumatra and Kalimantan are again changing; large severe fires can occur following brief dry spells and during non-ENSO years (Pittman *et al* 2013, Gaveau *et al* 2014). Moreover, an increasing proportion of these fires occur within peatlands (Miettinen *et al* 2011a, Marlier *et al* 2015).

Indonesian peatlands (~200 000 km^2) comprise ~5% of global peatland area and store an estimated 57 Gt carbon (C), ~9.4–12% of the global peat C pool (Page *et al* 2011). These ecosystems are highly sensitive to reductions in precipitation—especially extended ENSO-associated droughts (Field *et al* 2009)—that lower the water table, thereby leading to peat desiccation and enhancing peat flammability (Usup *et al* 2004, Wooster *et al* 2012). Due to high rates of forest conversion and peatland drainage associated with industrial-scale agricultural expansion (e.g., oil palm), peatlands have become increasingly vulnerable to fire (Hooijer *et al* 2006, Miettinen *et al* 2011a, Carlson *et al* 2013, Margono *et al* 2014, Turetsky *et al* 2015).

Peatland fires not only turn peatlands from C sink to source, but they also release large amounts of other gases and particles to the atmosphere; peat emissions are often orders of magnitude greater than those from other land-cover types (Christian *et al* 2003, Iinuma *et al* 2007, van der Werf *et al* 2010, Akagi *et al* 2011). For example, peat combustion is a significant source of biologically important nutrients, including nitrogen (N), phosphorus (P), sulfur (S), and potassium (K) (Andriesse 1988) that are often stored in peat for millennia (Weiss *et al* 2002). Emitted gases, particles, and chemicals are eventually delivered to downwind ecosystems, either dissolved in precipitation (wet or fog deposition) or directly in dry form (dry deposition; Weathers and Ponette-González 2011). After hitting the forest canopy, exchange via uptake and/or leaching can occur before nutrients are deposited to soils in throughfall (Ponette-González *et al* 2014), resulting in the redistribution of nutrients across the landscape (Ponette-González *et al* 2010a).

Ombrogenous peatlands are particularly sensitive to changes in atmospheric nutrient loading because they derive nutrients exclusively from the atmosphere and are also often nutrient limited (Tipping *et al* 2014). Research in boreal and temperate peatlands indicates that increases in atmospheric deposition can alter ecosystem C fluxes through effects on species composition, productivity, decomposition, and CH_4 flux (Gauci *et al* 2004, Bragazza *et al* 2006, Limpens *et al* 2008, Frolking *et al* 2011).

Given the magnitude of peatland fire emissions and the potential consequences of resultant deposition for biogeochemical cycling, here we address the following question: what is the influence of fires on biogeochemical fluxes of three biologically important plant nutrients (N, P, S) to adjacent, nutrient-limited peatland ecosystems? Specifically, what is the magnitude of fire-related N, P, and S fluxes to peat soils and what are the most important sources of N, P, and S in throughfall?

2. Methods

2.1. Study site and climate

This study was conducted in Kubu Raya District, West Kalimantan, Indonesia, ~35 km from the coast in a 12 ha intact forested peatland (mean peat depth 3.5 ± 0.1 m (±SE), range 2.6 –5.4 m; 0 °12′55″ S, 109 °25′38″ E). Based on vegetation mapping and monitoring from 2005–2014, the density of woody stems >10 cm dbh is ~458 stems ha^{-1}, while mean leaf area index (LAI) under closed canopy is 3.4 ± 0.69 m^2 m^{-2}. Mean tree basal area is 15 ± 0.1 m^2 ha^{-1}, with aboveground biomass of 148 ± 4.0 Mg ha^{-1}. Large-scale oil palm plantations, degraded forest fragments, government-sponsored transmigration areas, open burned abandoned areas, and smallholder agricultural fields surrounded the sampling area in 2015 (figure 1(b)).

Climatic data compiled from Supadio International Airport in Pontianak (8 km from site) indicate that mean annual rainfall (1960–2014) is 3197 ± 481 mm (±SD), with considerable intra- and inter-annual precipitation variability due to local land-sea breezes, seasonal monsoons, and ENSO events (Qian *et al* 2013). Total annual rainfall ranges nearly two-fold, 2329–4912 mm, from strong dry El Niño to strong wet La Niña years. Monthly rainfall is highest during the onset of the northeast monsoon (Oct–Dec: 314–359 mm mo^{-1}) and lowest when the southwest monsoon prevails (Jun–Sep: 186–250 mm mo^{-1}). Fire events mirror these rainfall patterns with few fires recorded during wet months and peak burning during drier periods (Vadrevu *et al* 2015).

2.2. Fire, land use, and air quality observations

The MODIS Active Fire Product (Giglio *et al* 2003) was used to determine fire locations within 20 and 100 km radius buffers surrounding the forested peatland. Fire counts are likely underestimated with these MODIS

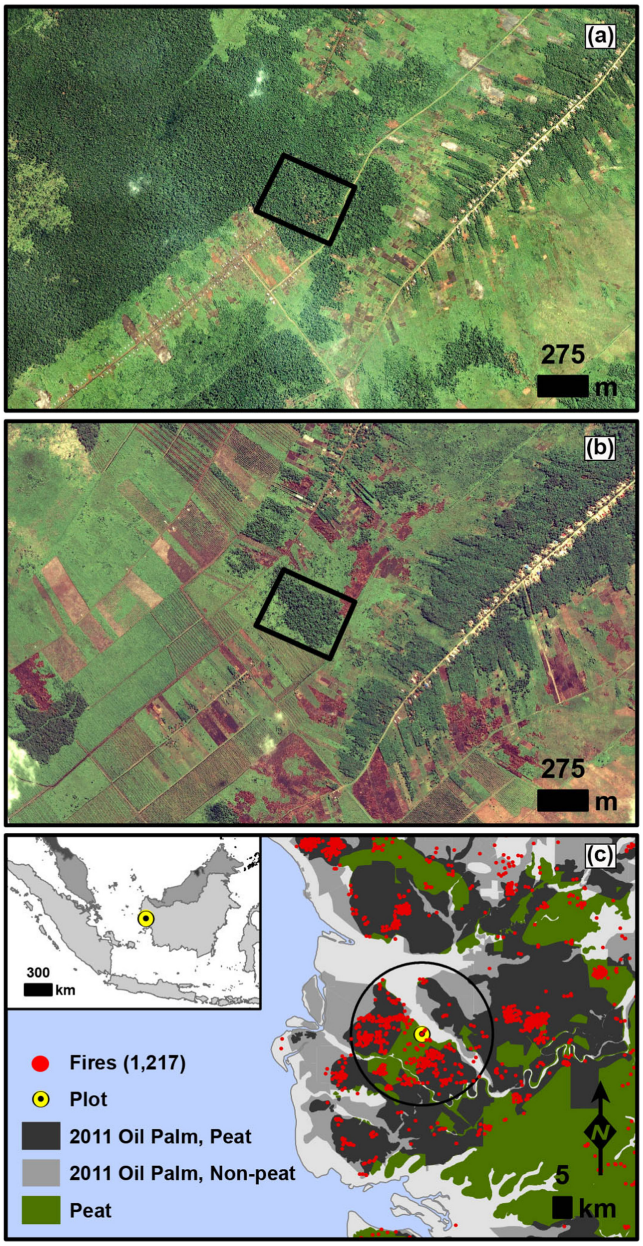

Figure 1. Dramatic land conversion around the 12 ha intact forested peatland (black box) from (a) June 2004 to (b) July 2015 in West Kalimantan, Indonesia (map data: Google, DigitalGlobe). Burning and clearing isolated the site's remaining forested peatlands. (c) Within 20 km of the site (black circle), fires detected from July 2013 to September 2014 were concentrated within oil palm leases on peatlands, which contained 78% of fires but only 49% of land area. Inset: location of study site in West Kalimantan, Indonesia.

data because clouds and smoke as well as forests with high LAI obscure fire detection (Roy *et al* 2008). Moreover, smoldering fires typical of peatlands are impossible to discern from flaming fires based on MODIS data, and are likely to be undetectable if they occur belowground (Elvidge *et al* 2015). Within 100 km surrounding this site, we digitized provincial and district agricultural oil palm lease records. Using peatland distribution information from RePPProT (1990) and Wetlands International (Wahyunto and Subagjo 2004), we then compiled fire hotspots by: (1) oil palm lease versus non oil palm lease; and, (2) soil type (i.e., mineral or peatlands; figure 1(c)). Daily observations of particulate matter concentration $<10\ \mu$m diameter (PM_{10}) were obtained from the government agency Badan Lingkungan Hidup Kota Pontianak (23 km from site).

2.3. Throughfall chemical fluxes

Across the 12 ha forested peatland, we measured dissolved total N, NO_3^-–N, NH_4^+–N, total P, PO_4^{3-}–P, SO_4^{2-}–S, and Cl^- in throughfall, water that flows through plant canopies and carries nutrients and pollutants from atmospheric deposition and canopy processing to soils. Using a spherical densiometer, the study site was stratified by canopy cover (0%–35% open, 35%–65% intermediate, 65%–100% closed). Six throughfall collectors were established randomly

within each canopy cover class, for 18 total collectors (figure S1). Three bulk rainfall deposition collectors (i.e., collectors that remain open between sampling events) were established in an adjacent clearing without canopy cover. Rainfall volume was recorded every five minutes with a 20 cm diameter tipping bucket rain gauge (Rainwise, Rainew 111) connected to a HOBO event logger (Onset, UA-003-64).

Throughfall and bulk deposition collectors were constructed with a 15 cm diameter funnel set on a PVC tube 1 m aboveground (after Weathers *et al* 2006, Ponette-González *et al* 2010b). Funnels were rinsed with deionized water and connected to plastic tubing, which drained into a 5 l plastic jug. A polywool filter was placed inside the funnel to prevent sample contamination. Water samples were collected weekly from July 2013 to September 2014. After each sample week (i.e., event), water volume was measured and a 300 ml aliquot was collected in a plastic bottle, wrapped in aluminum foil, and immediately frozen. Frozen samples were shipped to the Indonesian Institute of Sciences Limnology Laboratory in Bogor, Indonesia, where they were analyzed for total N (inorganic + organic N), NO_3^- –N, NH_4^+ –N, total P (inorganic + organic P), PO_4^{3-} –P, SO_4^{2-} –S, and Cl^- following standard protocols (APHA 1975, 2005, 2012, text S1).

Organic N was estimated as the difference between total N and dissolved inorganic N (DIN, NO_3^- – N + NH_4^+ –N) and organic P as the difference between total P and PO_4^{3-} –P. Chloride was used to determine the seasalt- SO_4^{2-} (ss − SO_4^{2-}) fraction in bulk rainfall using the average SO_4^{2-}/Cl^- ratio in seawater (0.14, Kroopnick 1977). Non-seasalt SO_4^{2-} (nss–SO_4^{2-}), an indicator of pollution, was calculated as the difference between SO_4^{2-} and ss–SO_4^{2-}. Volume-weighted mean (VWM) bulk rainfall and throughfall concentrations were computed for each sample event (i.e., week) or overall:

$$\mathrm{VWM} = \Sigma(\mathrm{conc}_i{*}\mathrm{precip}_i)/\Sigma\mathrm{precip}_i \qquad (1)$$

where i is the collector, conc is the solute concentration (mg l^{-1}) and precip is the rainfall or throughfall amount (mm). Bulk rainfall deposition and throughfall chemical fluxes for each event were then calculated by multiplying VWM concentrations by water volume (de Souza *et al* 2015). To compute the weighted mean and standard deviation of constituent concentrations across all weeks, as well as post-fire and normal weeks, we used the SDMTools package in R (VanDerWal *et al* 2014). From weekly VWMs, we calculated overall mean and standard deviation, weighting by the total volume across all collectors each week. We applied weighted least squares regression to determine whether concentrations between post-fire and normal weeks were significantly different.

2.4. Chemical signatures and sources

Hierarchical cluster analysis was used to identify groups of throughfall events with similar chemical composition. Prior to analysis, non-normally distributed data were log-transformed and all data were standardized by subtracting the mean and dividing by the standard deviation of each variable (Templ *et al* 2008). Cluster analysis was performed on NO_3^- – N, NH_4^+ –N, organic N, PO_4^{3-} –P, SO_4^{2-} –S, and Cl^- concentrations using Ward's clustering method. Organic P was correlated with PO_4^{3-} –P and thus excluded to minimize redundancy. After clustering, sources of elements in throughfall were explored using various methods. We examined between-cluster differences in molar ratios (N/P, N/SO_4^{2-}, SO_4^{2-}/NO_3^-, NH_4^+/NO_3^-, SO_4^{2-}/PO_4^{3-}, and SO_4^{2-}/Cl^-) using Kruskal–Wallis tests with post-hoc Steel Dwaas comparisons and within-cluster Pearson correlation coefficients among variables. Significance was set at $p < 0.05$. Analyses were performed using JMP v12 (SAS Institute, Cary, NC, USA). We also applied NOAA's Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT; Draxler and Rolph 2015) model to compute 168-h backward air mass trajectories for each throughfall event. Trajectories were computed at the surface (100 m), within the boundary layer (500 m), and above the boundary layer (1500 m).

3. Results

3.1. Dry periods and fire

During the 15 month (65 week) study (July 2013–September 2014), total rainfall was 2819 mm (188 mm mo^{-1}), well below the long-term monthly average for this region (266 mm mo^{-1}). Six dry periods occurred that varied in duration and severity (figure 2(a), table S1). These dry periods were associated with fire pulses—defined here as two or more weeks with ⩾10 fire hotspots detected within 20 km of the site—that also differed in magnitude, proximity to the focal site, and land-cover source (figure 2(b), table S1).

Total hotspots ranged 20-fold (10–196 hotspots) within 20 km of the site, and seven-fold (138–968 hotspots) within 100 km of the site. Approximately 80% of fires (20 km) and 53% of fires (100 km) were detected within oil palm leases, which covered 64% (20 km) and 49% (100 km) of the land area. A disproportionate number of fires was also detected within peatlands: 95% of fires versus 65% of land area within 20 km and 76% of fires versus 42% of land area within 100 km of the site. Seventy-eight percent (20 km) and 40% (100 km) of fires were located within oil palm leases on peatlands. PM_{10} levels closely tracked the fire pulses and exceeded 150 $\mu g\ m^{-3}$, the Indonesian daily ambient air quality standard, during all fire events

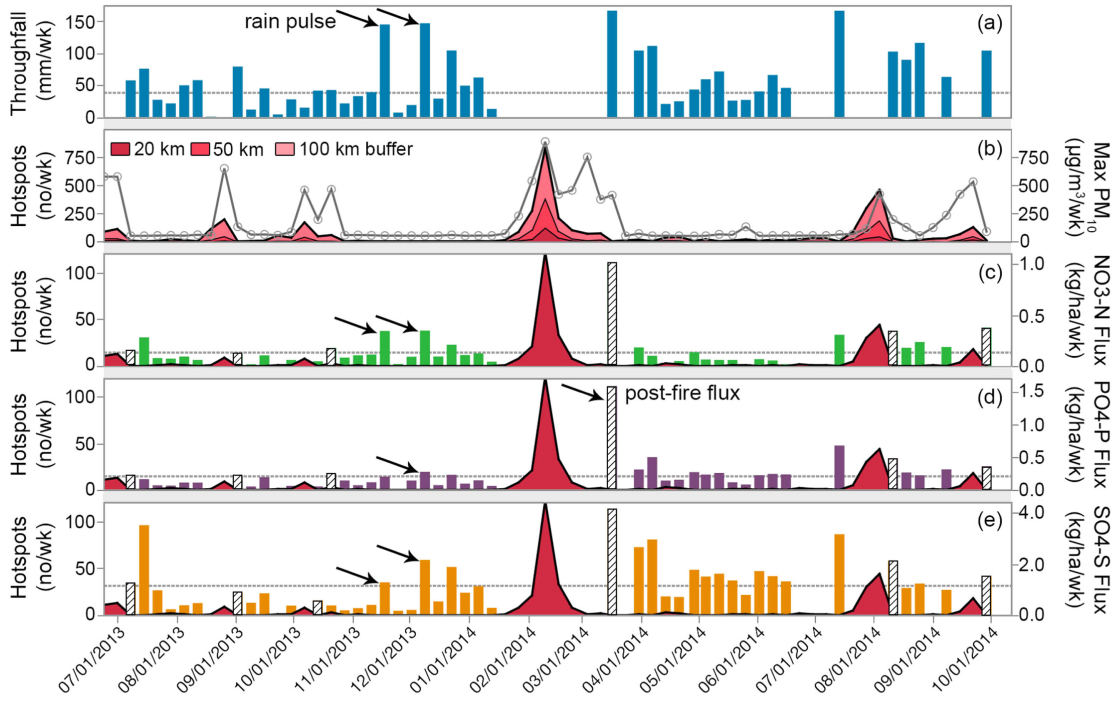

Figure 2. Throughfall N, P, and S fluxes (kg ha^{-1} wk^{-1}) to a tropical forested peatland in West Kalimantan, Indonesia from July 2013 to September 2014. (a) Weekly throughfall water flux (mm); (b) MODIS-detected fires within 20, 50, and 100 km radius buffers surrounding the 12 ha forested peatland and maximum weekly PM_{10} concentrations (grey line); (c) local fires ($\leqslant$20 km from the site) and weekly NO_3^-–N fluxes; (d) local fires and weekly PO_4^{3-}–P fluxes; and, (e) local fires and weekly SO_4^{2-}–S fluxes. Hatched bars indicate post-fire throughfall events ($n = 6$; table S1). Horizontal dashed lines indicate mean fluxes over the sample period and black arrows illustrate rain-driven enhancements in chemical fluxes.

(figure 2(b)). Daily PM_{10} values ranged from 194–886 μg m^{-3}, with 15 d > 301 μg m^{-3}.

3.2. Throughfall N, P, and S fluxes

From July 2013 to September 2014, a total of 44 throughfall events (i.e., weeks) were sampled. Average VWM concentrations of all solutes measured in throughfall over the study period were significantly greater than in rainwater ($p < 0.005$, Wilcoxon signed-rank test). Orthophosphate–P showed the most dramatic enrichment in throughfall concentration. Concentrations of PO_4^{3-}–P were 35-fold higher, while total P and organic P were 13-fold and four-fold higher, respectively, in throughfall than in bulk rainfall (table S2). For total N, NO_3^-–N, NH_4^+–N, DIN, and SO_4^{2-}–S, throughfall to bulk rainfall enrichment ratios ranged 1.4–2.3, indicating increasing concentrations with passage of water through the forest canopy.

Throughfall water flux was 95% of incoming rainfall (2688 mm). Notably, total P input, which averaged 0.024 kg P^{-1} ha^{-1} wk^{-1} in bulk rainfall (table S2), was 12-fold higher in throughfall, 0.29 kg P^{-1} ha^{-1} wk^{-1} (table 1). Dry deposition and/or canopy leaching (throughfall—bulk deposition) comprised 92% of the total P, 97% of the PO_4^{3-}–P and 76% of the organic P flux to the soil. The form of P delivered in rainwater differed markedly from that in below-canopy throughfall. Orthophosphate–P increased from ~33% of total P in rainwater to >78% of total P in throughfall, indicating that PO_4^{3-}–P is deposited primarily in dry form and/or that organic P is converted to PO_4^{3-}–P after deposition (Tipping *et al* 2014).

Total throughfall NO_3^-–N and NH_4^+–N fluxes were 117% and 110% higher (table 1), respectively, than bulk rainfall deposition of these ions (table S2). On average, mean DIN flux to soils was 0.25 kg ha^{-1} wk^{-1}. Differences between throughfall flux and bulk deposition were greater for DIN (5.8 kg ha^{-1}/15 mos) than for organic N (4.7 kg ha^{-1}/15 mos), indicating higher dry deposition and canopy exchange of DIN than of organic N. However, organic N represented a greater fraction of total N than DIN in both bulk rainfall (69%, table S2) and throughfall flux (60%, table 1).

Total SO_4^{2-}–S flux was two-fold higher than total N flux. Bulk rain S deposition was 31 kg ha^{-1}/15 mos, of which 45% was nss–SO_4^{2-}–S. An additional 22 kg S ha^{-1}/15 mos were delivered in throughfall, resulting in a total flux of 53 kg S ha^{-1}/15 mos to peatland soils (table 1). Assuming a similar proportion of nss–SO_4^{2-}–S in throughfall as in rainfall, 24 kg nss–SO_4^{2-}–S ha^{-1}/15 mos were deposited to this site.

3.3. Contribution of fires to nutrient loading

VWM concentrations and fluxes were calculated separately for the six post-fire throughfall events (14% of all 44 events) and normal events (table 1). Concentrations of inorganic N and P in throughfall were most

Table 1. Volume-weighted mean (VWM) throughfall concentrations (mg l^{-1}) and throughfall fluxes to soils for post-fire throughfall events ($n = 6$, preceding weeks $\geqslant$10 fires within 20 km buffer followed by rain), normal events ($n = 38$), and all events sampled from July 2013 to September 2014 in a rainfed forested peatland in West Kalimantan, Indonesia. Contribution of post-fire and normal events to the total throughfall flux is shown over the entire 15 month sampling period. Means $\pm$ SD are shown for VWM. Asterisk (*) indicates significant differences in VWM between post-fire throughfall and normal events ($p < 0.05$). Throughfall fluxes may not add due to rounding.

	Throughfall (mm)	VWM (mg l^{-1})			Throughfall flux (kg ha^{-1})			Contribution to total flux (15 mos, %)	
	All events	Total post-fire	Total Normal	Total all events	Total post-fire	Total normal	Total all events	Post-fire	Normal
Total N	2688	1.2 ± 0.39	0.95 ± 0.36	0.99 ± 0.37	6.7	21	27	25	75
DIN	2688	0.59 ± 0.29*	0.35 ± 0.17*	0.39 ± 0.21	3.5	7.5	11	32	68
Ammonium–N	2688	0.22 ± 0.22	0.15 ± 0.15	0.16 ± 0.16	1.3	3.1	4.4	29	71
Nitrate–N	2688	0.37 ± 0.15*	0.20 ± 0.080*	0.23 ± 0.12	2.2	4.3	6.5	34	66
Organic N	2688	0.59 ± 0.19	0.60 ± 0.28	0.60 ± 0.27	3.3	13	16	20	80
Total P	2688	0.64 ± 0.34*	0.42 ± 0.20*	0.46 ± 0.24	4.0	8.7	13	31	69
PO_4^{-3} –P	2688	0.50 ± 0.26*	0.33 ± 0.14*	0.36 ± 0.18	3.1	6.8	9.9	31	69
Organic P	2688	0.14 ± 0.09	0.09 ± 0.07	0.10 ± 0.08	0.9	1.9	2.8	31	69
Sulfate–S	2688	1.8 ± 0.58	2.0 ± 1.0	2.0 ± 0.97	10	43	53	19	81

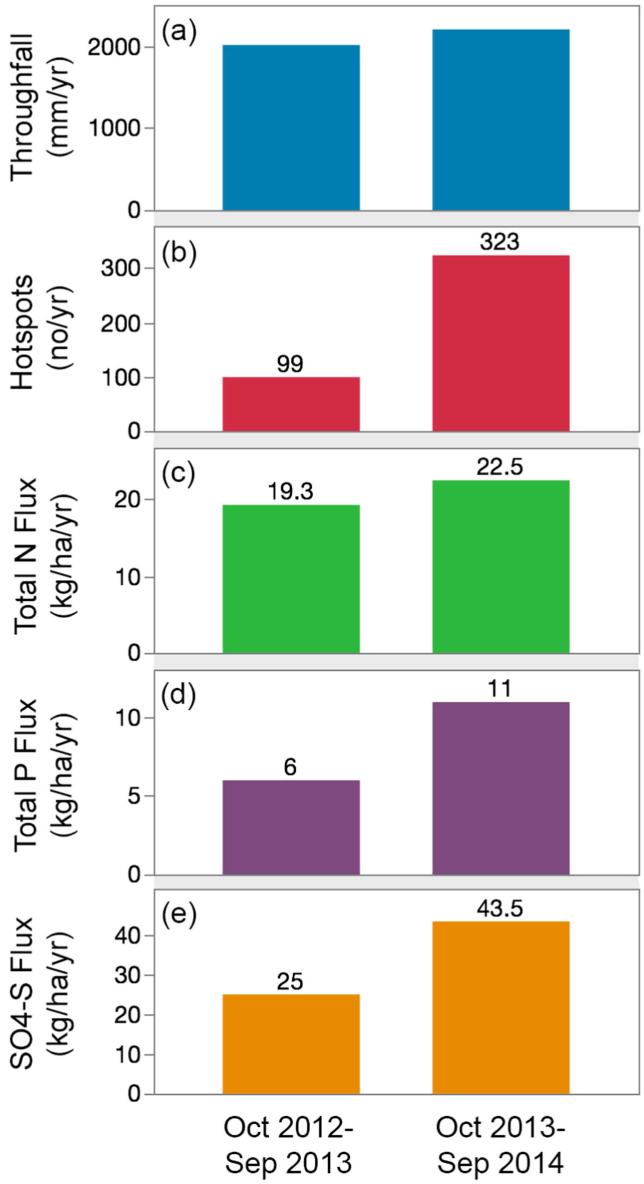

Figure 3. Total N, total P, and SO_4^{2-}–S fluxes in throughfall sampled during a high-fire (October 2012–September 2013) and low-fire (October 2013–September 2014) year in a forested peatland in West Kalimantan, Indonesia. (a) Total throughfall water flux (mm yr^{-1}); (b) total fire detections (⩽20 km of the site); (c) total N flux (kg ha^{-1} yr^{-1}); (d) total P flux (kg $ha^{-1}yr^{-1}$); and, (e) total SO_4^{2-}–S flux (kg $ha^{-1}yr^{-1}$).

elevated after fire, whereas organic N concentrations were similar between post-fire throughfall and all other events. Only SO_4^{2-}–S concentration was lower, on average, in post-fire throughfall compared to the other events.

Nineteen to 34% of the total throughfall chemical flux over the 15 month period was accounted for by six post-fire events (table 1). During these events, over a third of 'biologically-available' nutrient deposition occurred: PO_4^{3-} –P (31%), NO_3^- –N (34%), and NH_4^+ –N (29%). Post-fire events contributed 19% of total throughfall S and an estimated 19% of nss − SO_4^{2-}–S flux.

We also compared total N, total P, and S fluxes sampled in throughfall during two water years (October 2012–September 2013, October 2013–September 2014; text S2) with similar rainfall but a three-fold difference in total annual fire counts ⩽20 km from the site (figure 3). Total P and SO_4^{2-} –S in throughfall were two-fold higher, while total N was only slightly greater, during the high-fire compared to the low-fire year (figure 3). The slight increase in total N likely reflects the prevalence of organic N in throughfall, which was little affected by fire (table 1).

3.4. Chemical signatures and sources

Cluster analysis discerned five unique throughfall event clusters (table 2). Cluster 1 had the lowest NH_4^+/NO_3^- ratio and the highest SO_4^{2-}/Cl^- ratio. Throughfall SO_4^{2-} and organic P concentrations were significantly and positively correlated ($r = 0.62$, $p = 0.043$), suggesting a biomass source. Cluster 1

Table 2. Mean molar ratios in throughfall for five clusters of throughfall events (n) and bulk precipitation sampled from July 2013 to September 2014 in West Kalimantan, Indonesia. Letters indicate significant differences in ratios among clusters ($p < 0.05$).

Cluster	n	N/P	N/SO_4^{2-}	SO_4^{2-}/NO_3^-	NH_4^+/NO_3^-	SO_4^{2-}/PO_4^{3-}	SO_4^{2-}/Cl^-
1	11	5.1^{B}	1.2^{AB}	3.3^{B}	0.29^{B}	7.3^{A}	0.22^{A}
2	7	3.8^{B}	0.75^{B}	14^{A}	2.3^{A}	6.4^{AB}	0.20^{A}
3	9	9.6^{A}	2.2^{A}	2.9^{B}	0.71^{AB}	6.1^{A}	0.053^{B}
4	12	6.1^{AB}	1.7^{AB}	6.5^{AB}	1.6^{A}	7.0^{A}	0.17^{A}
5	4	4.6^{AB}	3.6^{AB}	1.2^{C}	0.87^{AB}	1.7^{B}	0.036^{B}
Bulk	44	52	1.4	6.0	0.98	151	0.11

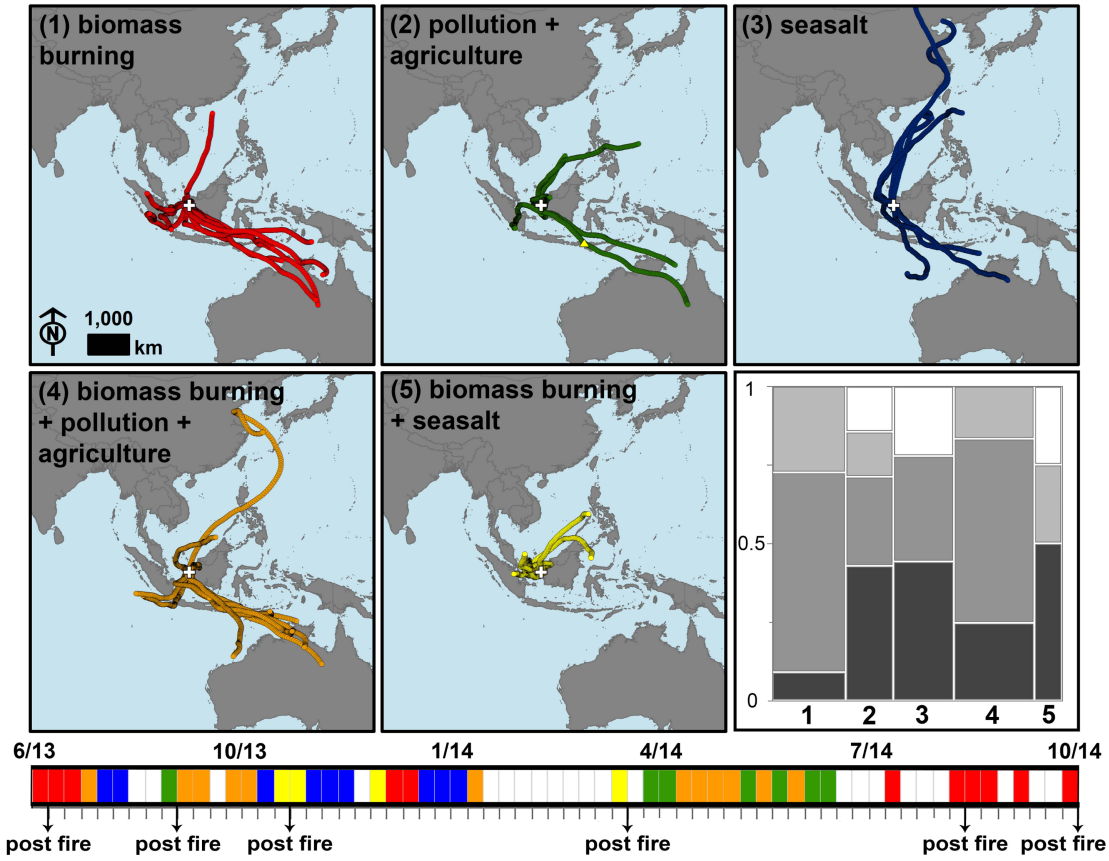

Figure 4. Dominant sources of N, P, and S in throughfall events sampled from July 2013 to September 2014 in West Kalimantan, Indonesia. Panels show air mass trajectories (500 m) for five clusters of throughfall events dominated by: (1) biomass burning; (2) pollution and agriculture (yellow triangle indicates location of Sangeang Api volcano); (3) seasalt; (4) biomass burning, pollution, and agriculture; and, (5) biomass burning and seasalt. Bottom right: prevailing winds for each cluster, including NE (dark grey), SE (medium grey), W/SW (light grey), and other (white). Along the x-axis, bar width represents the proportion of observations within each cluster.

contained three of six post-fire throughfall events, with most samples collected during peak biomass burning months in Indonesia and northern Australia (July–September; figure 4). Backward trajectories confirmed prevailing southeasterly winds that originated in northern Australia and then passed over southern Kalimantan before arriving at the focal site.

For Cluster 2, the mean SO_4^{2-}/NO_3^- ratio was as much as 14-fold higher compared to the other clusters (table 2), and nss–SO_4^{2-} comprised 79% of SO_4^{2-} in rainwater. Volcanic eruptions occurred in Southeast Asia during five of seven throughfall events included in this cluster (Global Volcanism Program 2013). However, SO_2 mass over Java and western Borneo was elevated during only two of these events, which followed the eruption of Sangeang Api in May 2014, coinciding with southeasterly airflow (NASA's Ozone Monitoring Instrument image database http://so2.gsfc.nasa.gov/). Cluster 2 also had the highest NH_4^+/NO_3^- ratio, an indicator of an agricultural N source. These results suggest that volcanic and anthropogenic pollution as well as agriculture were the dominant sources in Cluster 2 throughfall.

The SO_4^{2-}/Cl^- ratio in Cluster 3 was most similar, albeit depleted, to that in rainwater, and SO_4^{2-} and Cl^- concentrations showed a strong positive

correlation ($r = 0.74$, $p = 0.024$). Most events in this cluster occurred during the wet season when northeasterly winds prevail. Regardless of wind direction, backward trajectories show that air masses lingered over the ocean before their arrival at the focal site (figure 4), indicative of a dominant marine source in throughfall.

Similar to Cluster 2, Cluster 4 had a mean NH_4^+/NO_3^- ratio >1 (table 2) and, as with Cluster 1, throughfall SO_4^{2-} concentrations were strongly positively correlated with organic P ($r = 0.72$, $p = 0.0086$). Sulfate concentrations were also correlated with PO_4^{3-} ($r = 0.75$, $p = 0.0053$). Cluster 4 throughfall events were primarily influenced by southeasterly winds and otherwise variable winds in January, April, and May (figure 4). All events were temporally subsequent to two major fire pulses, suggesting a mixture of burning (Cluster 1), pollution, and agricultural sources (Cluster 2).

Cluster 5 contained two of six post-fire throughfall events, including the largest post-fire event in March 2014, and was characterized by a low SO_4^{2-}/Cl^- ratio. Throughfall events were affected by northeasterly and northwesterly winds, whereby air masses lingered over the ocean. This cluster had the lowest SO_4^{2-}/PO_4^{3-} ratio and distances traveled by the air masses were notably less than the other clusters, suggesting that throughfall chemistry was influenced mainly by local biomass burning rather than by long-range sources.

4. Discussion

4.1. Forested peatlands receive high N, P, and S deposition loads

At our peatland site, the magnitude of N, P, and S fluxes was remarkably high compared to other tropical systems where these fluxes have been measured (Ponette-González *et al* in press, Das *et al* 2011). Of major significance, PO_4^{3-}–P flux in throughfall was 7.9 kg ha^{-1}yr^{-1}. Such high fluxes rarely have been documented, and only within lowland tropical forest (e.g., 11 kg ha^{-1}yr^{-1}, Venezuela; Jordan *et al* 1980). However, bulk rainfall PO_4^{3-}–P deposition levels fell within the range of values for open field sites around the world (e.g., 0.02–0.65 kg ha^{-1}yr^{-1}; Tipping *et al* 2014). Comparison of bulk rainfall (0.24 kg ha^{-1}yr^{-1}) and throughfall flux (7.9 kg ha^{-1} yr^{-1}) indicates that the vast majority (97%) of inorganic P deposited to peatland soils was the result of dry deposition, a finding similar to observations from mature Neotropical dry forests (Das *et al* 2011).

Although our measured throughfall flux of 8.7 kg DIN ha^{-1} yr^{-1} agrees well with modeled estimates of total (wet + fog + dry) DIN deposition for regions of Southeast Asia (4–10 kg N ha^{-1}yr^{-1}; Vet *et al* 2014a, 2014b), the proposed critical N load limit for tropical forests is 5–10 kg N ha^{-1}yr^{-1}. Negative effects on species diversity, soil N cycling, and streamwater are expected beyond this limit (Pardo *et al* 2011), especially on highly acidic peatland soils.

Total annual S flux to this peatland was also extraordinarily high, totaling 42 kg S ha^{-1} yr^{-1}, of which we estimated 19 kg ha^{-1} yr^{-1} was nss–SO_4^{2-}–S. Assuming SO_4^{2-} in throughfall is a robust index of total (wet + fog + dry) atmospheric deposition in this peatland site (Weathers *et al* 2006, Ponette-González *et al* 2010a), our sample site ranks among the regions with the highest predicted total S deposition globally (i.e., East Asia 20–50 kg S ha^{-1} yr^{-1}; Vet *et al* 2014a, 2014b). The S flux reported here is comparable to estimates for forests located in Southern China's 'urban acid islands', areas <67 km from large cities where acid deposition exceeds critical loads (Du *et al* 2015).

Major global shifts in geographic source areas and ecosystem sinks for biologically important nutrients, including N, P, and S have occurred over the past three decades (Fowler *et al* 2013, Vet *et al* 2014a, 2014b). Emissions inventories and long-term network data show reductions in SO_2 and NO_x in North America and Europe with concomitant declines in the deposition of S and oxidized N (JRC/PBL 2011, Lehmann and Gay 2011, Tørseth *et al* 2012, US EPA 2014). In contrast, emissions and deposition of these compounds are rising in many Asian regions due to rapid urbanization, industrialization, as well as agricultural intensification and expansion (Dentener *et al* 2006, Vet *et al* 2014a, Du *et al* 2015). Although empirical data on P deposition are patchy, Asian ecosystems may become the new P deposition 'hotspots' (Wang *et al* 2015). Our measurements highlight this geographic redistribution of nutrients and pollutants over Asian ecosystems (Dentener *et al* 2006, Vet *et al* 2014a, Wang *et al* 2015).

4.2. Fire drives the redistribution of N, P, and S

Here, we show that fires have a disproportionate influence on below-canopy loading of N, P, and S to forested peatland soils. More than 30% of the inorganic N and P and nearly 20% of the nss–SO_4^{2-}–S in throughfall was deposited following major local and regional fire pulses (table 1, figure 2), although these throughfall events comprised only 14% of total sampled events. Dissolved inorganic nitrogen, NO_3^-, and PO_4^{3-} were the solutes whose concentrations and fluxes were most enhanced in post-fire throughfall.

It is well established that tropical biomass burning influences local and regional atmospheric chemistry and deposition, particularly N and P (Lobert *et al* 1990, Mahowald *et al* 2005). For example, experimental and observational studies of Indonesian peat fires find that NO_3^- is a major component of peat smoke aerosol (See *et al* 2007), while a dominant product of smoldering peat is ammonia gas (NH_3; Christian *et al* 2003). Biomass burning is also an important source of

rainwater and aerosol P (Maenhaut *et al* 2002, Mahowald *et al* 2008). During major Indonesian biomass burning periods in 1997 and 2006, rainwater sampled in Singapore contained elevated concentrations of NO_3^-, NH_4^+, and PO_4^{3-} compared with non-biomass burning periods (Balasubramanian *et al* 1999, Sundarambal *et al* 2010). Hsu *et al* (2014) collected aerosols downwind of the June 2013 peat fires in Riau Province; concentrations of total and soluble P in these samples were four-fold higher than in aerosol samples influenced by East Asian dust and pollution outflows.

While we cannot evaluate the relative contribution of biomass burning versus dust to our measured P fluxes, we note that soil dust may contribute to P loading during fire events. Three potential sources include: (1) dust emissions from biomass burning (Andreae *et al* 1998); (2) long-range dust transport from arid and semi-arid regions in East Asia (Lin *et al* 2007); and, (3) long-range dust transport from Australia (McGowan and Clark 2008). We are unaware of any study that has quantified the contribution of fires to atmospheric mineral dust emissions in Indonesia. Dust outflows from East Asia are most prevalent during the northeasterly monsoon (December–March), when only one of the fire events in this study occurred, and their influence on the southern South China Sea is thought to be minimal (Lin *et al* 2007). However, dust transport from Lake Eyre, Australia, to Borneo (June–August) is coincident with the peak biomass burning period in Southeast Asia (McGowan and Clark 2008). Therefore, it is possible that some of the P we measured in post-fire throughfall was derived from dust.

Because several studies report elevated SO_4^{2-} in rainwater and aerosol during peat fires (Ikegami *et al* 2001, See *et al* 2007), we were surprised to find that S concentrations were not enhanced in post-fire throughfall events. Two potential explanations include high local or regional S emissions from non-peat sources, and variability in fire emissions chemistry. First, fossil fuel and volcanic S emissions may have masked the influence of peat burning on atmospheric S fluxes. In Pontianak, the provincial capital of West Kalimantan (23 km from site), oil combustion was found to be the most important source influencing aerosol chemistry (Maenhaut *et al* 2002). Second, atmospheric emissions from flaming and smoldering fires differ in their chemical composition (Lobert *et al* 1990). In this study, we used MODIS Active Fire data that cannot distinguish between flaming and smoldering fires (Elvidge *et al* 2015). Smoldering fires are the fire type associated with high S emissions. Therefore, our correlations between fire detections and throughfall S concentrations possibly reflect the chemistry of flaming rather than smoldering fires.

Around our study site, land use—particularly industrial-scale agriculture—was a dominant driver of peatland burning. Fires were concentrated within oil palm leases on peatlands within 20 km (78% of fires versus 49% of land area) and 100 km (40% of fires versus 19% of land area) of the focal site. Yet, our data suggest that long-range transport of fire emissions from southern Kalimantan and northern Australia also influenced deposition at this site (figure 4). Land clearing and burning for oil palm plantation development are the dominant fire source not only in our study region, but also across Kalimantan and Sumatra (Curran *et al* 2004, Miettinen *et al* 2011b, Carlson *et al* 2012, 2013, Gaveau *et al* 2014, Marlier *et al* 2015). Atmospheric emissions from these activities are projected to increase over the next decade (Carlson *et al* 2012, 2013, Miettinen *et al* 2012).

The enhanced N, P, and S fluxes we documented in association with agricultural peat fires during a non-ENSO year indicate that emissions from larger and more severe fires typical of strong ENSO years (figure S2) could result in dramatically increased deposition to adjacent ecosystems. Our results showed that a three-fold increase in detected fires resulted in a two-fold increase in annual total P and S flux to peatland soils (figure 3). We also captured the influence of a severe fire pulse from January to March 2014 (196 fires $\leqslant$20 km from the site) on throughfall fluxes (table S1). Following this single fire event, 13% of DIN, 16% of PO_4^{3-} –P, and 8% of SO_4^{2-} –S measured over the study period was delivered to peatland soils as throughfall flux (figure 2).

5. Conclusions

In this study, we show that biomass burning in tropical peatlands plays a largely unrecognized yet critical role in the redistribution of major limiting nutrients within tropical landscapes. Our estimates indicate that as much as 30% or more of the annual load of inorganic N and P to Bornean peatlands occurred following fire events and that fire-emission-deposition of S could rival that of industrial pollution during high-fire strong ENSO years. As such, biomass fires may have significant fertilizing or polluting effects on recipient ecosystems, thereby altering the contribution of peatlands to climate change (Frolking *et al* 2011). While the ecosystem effects of biomass burning deposition remain uncertain in tropical regions, the sheer magnitude of the fluxes measured here indicate that fire-driven nutrient redistribution may have unforeseen consequences across a diversity of ecosystems, including forested peatlands as well as agroecosystems, wetlands and streams, oligotrophic ocean waters, and coral reefs.

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