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Tropical land-cover change alters biogeochemical inputs to ecosystems in a Mexican montane landscape

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Abstract. In tropical regions, the effects of land-cover change on nutrient and pollutant inputs to ecosystems remain poorly documented and may be pronounced, especially in montane areas exposed to elevated atmospheric deposition. We examined atmospheric deposition and canopy interactions of sulfate-sulfur (SO_4^{2-}-S), chloride (Cl^-), and nitrate-nitrogen (NO_3^--N) in three extensive tropical montane land-cover types: clearings, forest, and coffee agroforest. Bulk and fog deposition to clearings was measured as well as throughfall (water that falls through plant canopies) ion fluxes in seven forest and five coffee sites. Sampling was conducted from 2005 to 2008 across two regions in the Sierra Madre Oriental, Veracruz, Mexico.

Annual throughfall fluxes to forest and coffee sites ranged over 6–27 kg SO_4^{2-}-S/ha, 12–69 kg Cl^-/ha, and 2–6 kg NO_3^--N/ha. Sulfate-S in forest and coffee throughfall was higher or similar to bulk S deposition measured in clearings. Throughfall Cl^- inputs, however, were consistently higher than Cl^- amounts deposited to cleared areas, with net Cl^- fluxes enhanced in evergreen coffee relative to semi-deciduous forest plots. Compared to bulk nitrate-N deposition, forest and coffee canopies retained 1–4 kg NO_3^--N/ha annually, reducing NO_3^--N inputs to soils. Overall, throughfall fluxes were similar to values reported for Neotropical sites influenced by anthropogenic emissions, while bulk S and N deposition were nine- and eightfold greater, respectively, than background wet deposition rates for remote tropical areas.

Our results demonstrate that land-cover type significantly alters the magnitude and spatial distribution of atmospheric inputs to tropical ecosystems, primarily through canopy-induced changes in fog and dry deposition. However, we found that land cover interacts with topography and climate in significant ways to produce spatially heterogeneous patterns of anion fluxes, and that these factors can converge to create deposition hotspots. For land managers, this finding suggests that there is potential to identify species and ecosystems at risk of excess and increasing deposition in montane watersheds undergoing rapid transformation. Our data further indicate that montane ecosystems are vulnerable to air pollution impacts in this and similar tropical regions downwind of urban, industrial, and agricultural emission sources.

Key words: air pollution; atmospheric deposition; chloride; cloud forest; critical loads; fog; global environmental change; ion-exchange resins; nitrogen; shade coffee; sulfur; throughfall.

Introduction

Increasing atmospheric deposition is a significant component of the suite of human-accelerated environmental changes occurring across the globe (Vitousek et al. 1997, Dentener et al. 2006, Galloway et al. 2008). Over the previous three decades, understanding of the causes and consequences, temporal trends, and spatial patterns of atmospheric deposition in temperate ecosystems has improved considerably (Likens and Bormann 1974, Johnson and Lindberg 1992, Ollinger et al. 1993, Lovett 1994, Fenn et al. 1998, Weathers and Lovett 1998, Weathers et al. 2000, 2006*a*, Driscoll et al. 2001, Neal et al. 2001, Holland et al. 2005). Yet, with the notable exceptions of Hawai'i (Chadwick et al. 1999, Vitousek 2004) and Puerto Rico (Heartsill-Scalley et al. 2007), a paucity of long-term, systematic, and standardized data collection across the majority of tropical regions has limited research on the role of atmospheric deposition in tropical biogeochemical cycles (Rodhe and Herrera 1988). Thus, while the geographic extent and ecological effects of enhanced deposition in tropical environments remain uncertain (Asner et al. 2001), variation in the sources and strength of emissions coupled with the characteristics of recipient ecosystems (i.e., species composition, nutrient limitation) suggest that deposition patterns and ecosystem responses may

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contrast greatly with those recorded in temperate areas (Tanner et al. 1998, Martinelli et al. 1999, Matson et al. 1999, Lawrence et al. 2007).

Some tropical mountains appear particularly vulnerable to high rates of atmospheric deposition because of distinct meteorological conditions, topographic effects on the deposition process, and proximity to natural and anthropogenic emissions (Weathers et al. 1988, Asbury et al. 1994, Eklund et al. 1997, Daly et al. 2007, Boy et al. 2008). Nutrients and pollutants are delivered to the Earth surface via wet, fog, and dry deposition (Lovett 1994). Wet deposition is a function of precipitation volume and chemistry, while fog and dry deposition are influenced by such factors as particle and gas concentrations, wind speed, relative humidity, canopy wetness, liquid water content, fog immersion time, and fog droplet size (Weathers et al. 2006*b*). In mountainous terrain, increased levels of orographic precipitation (rain, fog, and snow) and wind exposure contribute to enhanced deposition at high-elevation sites (Lovett and Kinsman 1990), as well as to windward-facing slopes, ridges, edges, and canopy gaps (Lindberg and Owens 1993, Weathers et al. 1995, 2000, 2001, 2006*a*, Fenn et al. 2003, Nanus et al. 2003, Ewing et al. 2009). Furthermore, fog may be at least as important as rain in delivering chemical substances to montane ecosystems (Lovett and Reiners 1986, Weathers et al. 1986, 1988, Carrillo et al. 2002). Ion concentrations in fog water are often severalfold higher than in rainwater (Weathers et al. 1986, 1988, Gordon et al. 1994, Clark et al. 1998*a*, Liu et al. 2005, Raja et al. 2005). Therefore, even relatively low fog water inputs may contribute substantially to total chemical deposition (Weathers et al. 2000, Ewing et al. 2009). Where mountain ranges occur in close proximity to oceans, volcanoes, urban/industrial, or agricultural areas, these sources interact synergistically to create new or exacerbate existing deposition hotspots (Daly and Wania 2005).

In addition to abiotic factors, variation in tree species composition and canopy structure have been shown to strongly influence rates and spatial patterns of deposition (van Ek and Draaijers 1994, Weathers et al. 2000, 2006*a*, De Schrijver et al. 2008). Plant canopies modify the chemistry of water inputs to soil in two ways: directly through canopy-exchange processes (i.e., leaching and uptake) and indirectly through interactions with particles and gases deposited to receptor surfaces via fog and/or dry deposition (Cape et al. 1987). In general, forests tend to be efficient scavengers of nutrients and pollutants due to their high surface roughness, resulting in elevated inputs to forests compared to shorter vegetation (e.g., crops, grassland; Fowler et al. 1999). Recent studies from mixed-use landscapes show that tropical tree-based land-use systems may also incur substantial loading. Lilienfein and Wilcke (2004) recorded a significant increase in hydrogen (H) and aluminum (Al) fluxes under pine plantations relative to savanna forest, degraded and productive pasture, and corn–soybean plots, while Schroth et al. (2001) found that throughfall phosphorous (P) and potassium (K) inputs to palm monocultures were significantly greater than to open areas. Tropical forests also retain significant quantities of elements in the canopy layer, through foliar absorption, retention in canopy organic matter, and uptake by canopy-dwelling organisms (Lang et al. 1976, Clark et al. 2005). Thus, disturbances that alter canopy structure and composition can impart considerable change in ecosystem nutrient cycling (Nadkarni et al. 2004).

In Latin America, Asia, and Africa, several tropical montane regions are concomitantly experiencing rapid and extensive land-cover change and increasing atmospheric deposition. To date, relatively few studies have examined how these processes, singly or combined, alter nutrient and pollutant inputs to ecosystems. Therefore, this study had three major objectives: (1) estimate total (wet + dry + fog) atmospheric deposition to three contrasting land-cover types (tropical montane forest, coffee agroforest, and clearings) using sulfate-S as a conservative chemical tracer; (2) examine the effects of land cover on canopy-level processing (i.e., leaching and uptake) of Cl^- and NO_3^--N; and, (3) assess deposition results within a broader regional context.

Methods

Study region

Central Veracruz is located along the eastern escarpment of the Sierra Madre Oriental mountain range in eastern Mexico (19°20′ N, 96°50′ W; Fig. 1). This orographic barrier separates the low-lying Gulf Coastal Plain from the upland Neovolcanic Plateau and is characterized by a broad altitudinal gradient that spans from sea level to the summits of Cofre de Perote (4282 m above sea level) and Pico de Orizaba volcanoes (5675 m) over a mere 100 km horizontal distance.

With prevailing tropical easterlies as a constant moisture source, the central Veracruz highlands experience relatively humid conditions year round (Jáuregui Ostos 2004). Within the mid-elevation belt (1000–2200 m), mean annual temperature decreases with elevation from 20° to 12°C. Total annual rainfall ranges from 1000 to 2200 mm/yr (García 1973). In addition, two discrete precipitation seasons occur: a wet season (May–October: Atlantic Hurricane season months) and a drier season (November–April). During this dry period, northers deliver moisture in the form of drizzle and fog. El Niño Southern Oscillation (ENSO) events enhance precipitation variability on interannual time scales, primarily by increasing the volume and shifting the temporal distribution of dry season rainfall (Cavazos 1997). During winter El Niño events, rain inputs to montane ecosystems (1000–1600 m) in central Veracruz may increase as much as 28% (Ponette-González et al. 2010).

Superimposed upon this environmental heterogeneity is a patchwork mosaic of forested, agricultural, and

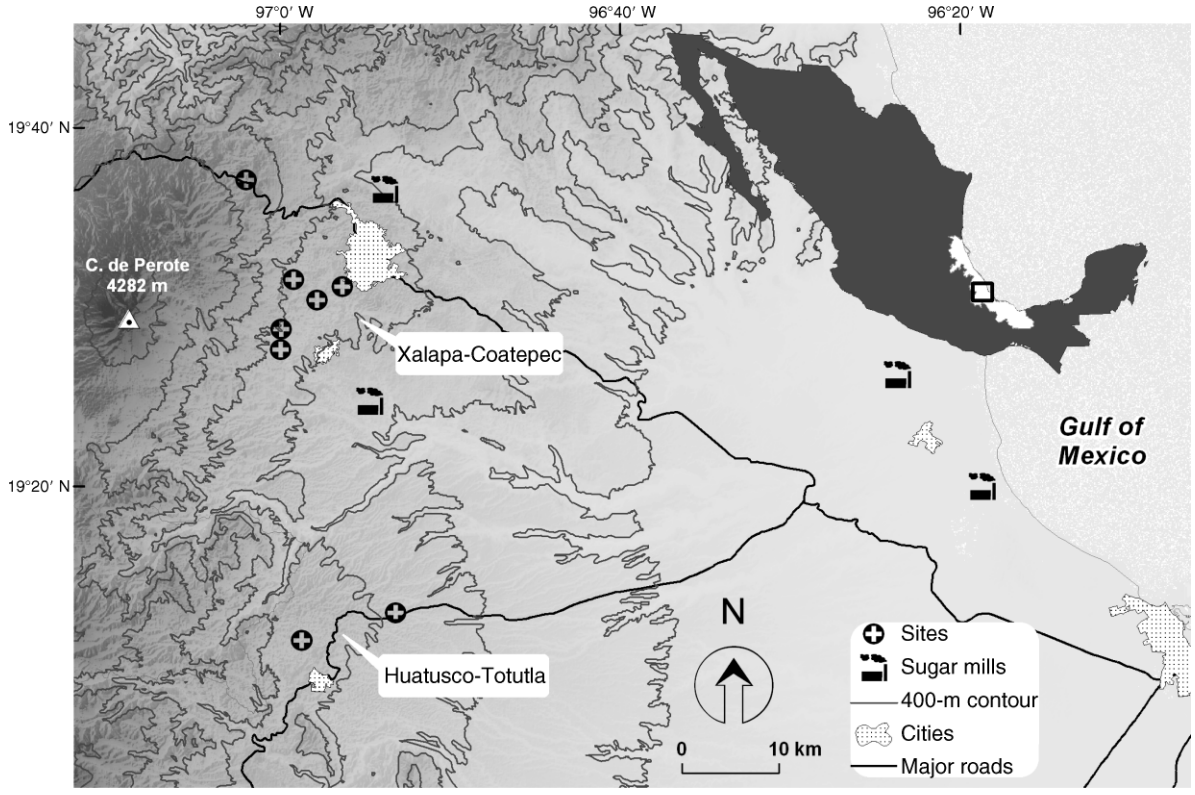

FIG. 1. Map of the study region in central Veracruz (black box), Mexico, showing study sites, sugar mills, cities, and major roads. Elevation increases from sea level (light gray) to the summit of Cofre de Perote volcano (4282 m; dark gray) over 100 km horizontal distance. After 2400 m, contour lines are every 800 m. Inset shows Veracruz State in eastern Mexico.

urban ecosystems with diverse land-use histories. At mid-elevations, lower (800–1800 m) and upper (1800–2700 m) montane forests contain a rare mixture of temperate and tropical elements, including 50 species of temperate plants related to northeastern U.S. flora (Graham 1999). Forest vegetation belts are characterized by distinct plant communities and species composition. Dominant trees in lower montane forest canopies include deciduous species such as *Carpinus caroliniana* (Betulaceae), *Liquidambar styraciflua* (Hamamelidaceae), and *Quercus* spp. (Fagaceae), whereas Neotropical evergreen genera, including *Cinnamomum effusum* (Lauraceae), *Hedyosmum mexicanum* (Chloranthaceae), *Oreopanax xalapensis* (Araliaceae), and *Turpinia insignis* (Staphylacaceae) thrive in the understory (Williams-Linera 2002). Above 1800 m, broadleaf trees are gradually replaced by conifers, such as *Pinus patula* (Pinaceae), *Pinus ayacahuite* (Pinaceae), and *Abies religiosa* (Pinaceae). Canopy epiphyte diversity, abundance, and biomass are generally high within these forest ecosystems (Hietz and Hietz-Seifert 1995).

Much of the original forest cover in central Veracruz has been cleared or severely degraded by human activity, causing forest fragments to vary greatly in vegetation structure and species composition. Often restricted to steep slopes and hilltops, residual forest patches are typically <40 ha in size with a few large contiguous blocks no greater than 900 ha (Williams-Linera et al. 2002). Forest fragments are frequently surrounded by shade coffee agroforests and cattle pastures. In general, small-scale coffee holdings (≤2 ha in size) are located on marginal lands (e.g., steep slopes; Trujillo 2008), while medium- and large-scale plantations exceed 100 ha and occupy sloping as well as flat areas. Management practices vary considerably, but most coffee is cultivated under the shade of evergreen fruit, leguminous, and timber trees. Common shade species include *Inga* spp. (Leguminosae), *Trema micrantha* (Ulmaceae), and *Enterolobium cyclocarpum* (Leguminosae). Many cattle pastures are cultivated with exotic grasses (e.g., *Cynodon plectostachyus*, stargrass) and encompass <10 ha when smaller to hundreds of hectares when larger landholdings. Along with sugarcane, these land-use systems occupy 40–60% of the land area in the study region (Muñoz-Villers and López-Blanco 2008). Sugarcane (*Saccharum officinarum*) fields are primarily smallholdings (<1–6 ha) but combined may cover large land areas (>1000 ha). Sugarcane is planted between September and March and burned and harvested from December to May. The most common soil types underlying these land uses are Andosols at mid- to upper elevations (≥1500 m)

TABLE 1. Description of study sites in central Veracruz, Mexico, where throughfall fluxes were sampled.

Site	Sample year	Region	Elevation (m above sea level)	Land cover	Rainfall (mm)	Soil type	Sensitivity class
A1	2005–2006	Xalapa-Coatepec	1450–1500	forest	1873	Andosol	3
A2	2005–2006	Xalapa-Coatepec	1400–1450	coffee	1873	Andosol	3
B3	2005–2006	Xalapa-Coatepec	1300–1350	forest	2376	Andosol	3
B4	2005–2006	Xalapa-Coatepec	1300–1350	coffee	2376	Andosol	3
C5	2005–2006	Xalapa-Coatepec	1450–1500	forest	2001	Andosol	3
C6	2005–2006	Xalapa-Coatepec	1350–1400	coffee	1668	Andosol	3
D7	2006–2007	Huatusco-Totutla	1050–1150	forest	1339	Acrisol	1
D8	2006–2007	Huatusco-Totutla	1050–1150	coffee	1339	Acrisol	1
E9	2006–2007	Huatusco-Totutla	1350–1400	forest	1788	Andosol	3
E10	2006–2007	Huatusco-Totutla	1350–1400	coffee	1582	Andosol	3
F11	2006–2007	Xalapa-Coatepec	1550–1600	forest	2421	Andosol	3
G12	2006–2007	Xalapa-Coatepec	2200–2250	forest	1105†	Andosol	3

Notes: Sites with the same capital letters indicate paired forest–coffee plots. Sites 11 and 12 were compared with cleared areas. Soils are classified according to Geissert and Ibañez (2008) and sensitivity to acidic deposition (Kuylenstierna et al. 2001): from 1 (most sensitive) to 5 (least sensitive).

† Rainfall not measured at this site. Data indicate annual normal for 1971–2000 (National Weather Service, ⟨http://smn.cna.gob.mx/⟩).

and Acrisols at lower elevations (≤1000 m; Rossignol et al. 1987).

Currently, expansion of the Xalapa metropolitan and surrounding peri-urban areas is driving conversion of forests and coffee agroforests for residential development and sugarcane production (see Plate 1). In addition, the central Veracruz highlands are downwind of Mexico's industrialized Gulf Coastal Plain, where the highest SO_2-emitting power plant in North America (Miller and Van Atten 2004), oil and gas extraction with refining facilities, petrochemical complexes, and industrial sugarcane plantations are all concentrated. These activities have been shown to alter atmospheric chemistry and deposition in this and other tropical regions (Báez et al. 1997, Matson et al. 1998, Lara et al. 2001).

Experimental design

This study was conducted in the Xalapa-Coatepec (XAC) and Huatusco-Totutla (HUT) regions (Fig. 1). Three distinctive land-cover types were chosen for comparison: montane forest, coffee agroforest (commercial polycultures, sensu Moguel and Toledo 1999), and cleared areas. Cleared areas include cattle pastures, areas with low grass, and other zones without canopy cover. To ensure that these land-cover types were exposed to similar meteorological conditions, we selected five forest fragments (3–93 ha) adjacent or near (1–3 km) shade coffee holdings (6–90 ha) and clearings. Two additional forest sites (4 ha, 29 ha) above the elevational limit of coffee cultivation (~1500 m) were compared to cleared areas. No agrochemicals (i.e., fertilizers, herbicides, or fungicides) were applied to coffee leaves or soils in these coffee sites throughout the duration of the study. A full description of site characteristics is provided in Table 1.

In mountainous landscapes, fine-scale topographic variability is one of several factors affecting atmospheric deposition (Weathers et al. 1992, 2000, 2006*a*, Ollinger et al. 1993). Because our goal was to discern the effects of land cover on chemical fluxes, we established plots on east-facing slopes between 20° and 30°, and, with the exception of one forest site (2200 m), between 1000 and 1600 m. We assumed that as a result of similar slope aspect and steepness, all tree crowns in forest and coffee sites would be equally exposed to prevailing winds, causing slope to act as a "functional" edge (Weathers et al. 2000), and, in turn, minimizing within-plot edge effects. In addition, we avoided sampling near sharp forest–pasture boundaries at two sites. Measurements were conducted ~20 m into the plot from this land-use edge, the approximate distance at which deposition rates have been shown to decrease by 50% (Weathers et al. 1992).

Throughfall water inputs

In a related study, we measured throughfall water (water that falls through gaps in plant canopies) inputs to these same sites. Between 14 and 16 throughfall samplers were established in ~1300-m^2 plots within six of the forest ($n = 90$) and all five shade coffee sites ($n = 62$). Twenty cm diameter polyethylene funnels were deployed, and each was attached to flexible Tygon tubing and set onto PVC pipe (Weathers et al. 2001, 2006*a*). A polywool filter was inserted into the neck of the funnel to prevent debris from blocking the flow of water. Samplers were mounted 1 m aboveground and spaced at 12-m fixed intervals. Seventeen "reference" rainfall collectors were installed in adjacent clearings. Water from throughfall and rainfall samplers was collected weekly from April 2005 to March 2008. Details on sampling methodology, statistical analyses, and results can be found in Ponette-González et al. (2010).

Bulk and fog deposition

Four paired bulk and passive fog deposition collectors along with a single bulk collector were installed in clearings ($n = 5$ bulk collectors, $n = 4$ fog collectors).

Bulk collectors accumulate primarily wet inputs, but also some proportion of dry particles. Passive fog samplers collect horizontal fog as well as some wind-driven rain and dry inputs. In December 2005, paired bulk and fog samplers were installed at two sites in the Xalapa-Coatepec region, and in November 2006, at an additional higher-elevation site (2200 m; Fig. 1). Two bulk plus one fog sampler were established in the Huatusco-Totutla region in November 2006. Bulk and fog deposition collectors were located on the periphery of towns and cities and distant from local industries. We considered these sites to be representative of regional atmospheric conditions.

Following the design of Simkin et al. (2004) and Weathers et al. (2006*a*), bulk samplers were constructed from a 20 cm diameter plastic high-density polyethylene (HDPE) funnel connected to flexible Tygon tubing, a plastic HDPE connector, and an anion-exchange resin column. Passive string fog samplers (i.e., "artificial" collectors) were constructed after Falconer and Falconer (1980). The cylinder-shaped collectors were made from polypropylene (25 × 50 cm) and were strung with 0.5 mm diameter Teflon strand. Each collector was connected to a 31 cm diameter plastic HDPE funnel and an anion-exchange resin column, and then covered with a 50 cm diameter hood to exclude vertical rainwater inputs. During fog events, fog water droplets impacted onto Teflon strands, dripped through the funnel and anion-exchange resin column. Anions in bulk and fog water adhered to positively charged exchange sites on resin.

Throughfall chemistry

We also measured the chemical composition of throughfall, water that flows through plant canopies and carries nutrients and pollutants from atmospheric deposition and canopy processing to soils. With biologically conservative chemical tracers, such as SO_4^{2-}-S, throughfall has been successfully employed to estimate rates of total atmospheric deposition (wet, dry, and fog) to forests (Johnson and Lindberg 1992, Weathers et al. 1995, 2000, Simkin et al. 2004). Relationships between atmospheric SO_4^{2-} and SO_2 deposition and total SO_4^{2-} measured in throughfall have been well established (Johnson and Lindberg 1992, Lindberg et al. 1992), suggesting that relative to total SO_4^{2-} deposition, canopy uptake and foliar leaching of S are minimal (<15% of total deposition; Lindberg and Garten 1988, Lindberg and Lovett 1992). Together, measurements of bulk deposition (ions that have not interacted with canopies) and total throughfall inputs of conservative ions can thus be used to estimate fog plus dry deposition fluxes to soils. Total throughfall flux of biologically active ions (i.e., ions retained in or leached from plant canopies) such as NO_3^--N and NH_4^+, while providing a robust measure of total flux to soils, often cannot be interpreted as an estimate of total deposition to plant canopies (Weathers et al. 2006*a*).

Using anion-exchange resin throughfall collectors (Simkin et al. 2004, Weathers et al. 2006*a*), throughfall SO_4^{2-}-S, Cl^-, and NO_3^--N fluxes were measured under the canopies of three forests and three coffee agroforests in the XAC region from 13 October 2005 to 12 October 2006, and under four forests and two coffee agroforests in XAC and HUT from 13 November 2006 to 12 November 2007 (Table 1). A total of 122 anion-exchange resin throughfall collectors were installed at these sites. Identical to bulk deposition samplers, 10 resin throughfall samplers were established within each site along five separate transects using a stratified-random sampling design. Collectors were installed >20 m from the land-use boundary at sites A1 and E11, 1 m aboveground, and spaced at 12-m intervals. A polywool filter was inserted into the neck of each funnel to prevent debris from inhibiting water flow. As resin samples are more chemically stable than anions in solution (Simkin et al. 2004), resin columns were collected every 4–6 weeks during the wet season, and every 6–10 weeks during the dry season, totaling seven sampling periods per year. In addition, in situ bulk and throughfall water pH measurements were recorded monthly from October 2006 to August 2007 with a 20 digital pH/mV/ORP meter (Barnant, Barrington, Illinois, USA).

Field sampling

Field sampling materials were prepared at the Functional Ecology Laboratory at the Institute of Ecology, Asociación Civil, in Xalapa, Veracruz. Before sampling, bulk, fog, and throughfall collector components and polywool filters were washed, soaked in deionized water for 24 hours, and then rinsed with deionized water. After drying in clean enclosed areas, components were assembled and packed individually in large plastic bags. Anion-exchange resin columns were prepared after Simkin et al. (2004) and Weathers et al. (2006*a*). Briefly, Dowex Monosphere 550-A (OH^- form) anion-exchange resin (Dow Chemical Company, Midland, Michigan, USA) was used. The resin was rinsed with deionized water to remove excess NaOH and broken resin beads. The 0.64 cm-thick 30-μ pore-size filter in each chromatograph column was replaced with a 0.32 cm-thick 30-μ pore-size filter to reduce clogging. Each column was filled with 20 mL of anion-exchange resin. Prepared columns were then refrigerated before use.

Funnels and anion-exchange resin columns were installed at the onset of the 2005 and 2006 dry seasons. As distance between sites did not permit all columns to be set on the same day, funnels and resin columns were set over the course of a 32-hour rain free period. Resin columns were collected and new columns replaced during 32-hour rain free periods. Immediately after each sample collection, funnels were cleaned in the field with deionized water and new clean collector components attached along with new polywool filters.

Extraction and sample analysis

Samples were shipped to the Cary Institute of Ecosystem Studies for extraction and analysis. A total of 945 anion-exchange resin columns were extracted in the laboratory following the methods of Simkin et al. (2004) and Weathers et al. (2006*a*). Field resin samples plus two "blank samples" per sample period ($n = 28$) were extracted three times in 1.0 mol/L potassium iodide (KI). Samples were then diluted in double deionized water. A Dionex DX-500 ion chromatograph (IC; Dionex Corporation, Sunnyvale, California, USA) fitted with a Dionex Ion-Pac AG9-HC guard column and an AS9-HC analytical column were used to analyze the samples. The detection limit of this method is 0.02 mg/L. A NIST-traceable known, 11 standards-as-samples and four replicate samples were included with each instrument run for quality assurance-quality control (QA/QC). QA/QC results fell within ±5% accuracy.

Values below the detection limit were set to half the detection limit. Concentrations of SO_4^{2-}, Cl^-, and NO_3^- were then converted to fluxes per element (kg/ha) per sampling period by incorporating collector surface area. Bulk collector surface area was 324 cm^2. Fog collector surface area was 1069 cm^2 and was calculated based on the area of a cylinder.

Statistical analysis

"Water year," the sum of one wet and one dry season, is used to report annual throughfall flux and deposition data (±SE). The net throughfall flux was calculated for all anions:

$$TF - BD = NTF \quad (1)$$

where TF is the throughfall flux (kg element/ha), BD is bulk deposition (kg/ha) of the nearest sampler, and NTF is the net throughfall flux (kg/ha). Here, we use negative net throughfall values (NTF $<$ 0) to indicate canopy uptake, whereas positive net throughfall values (NTF $>$ 0) indicate fog/dry deposition and/or canopy leaching. In our calculations of mean flux per land-cover type, we used only data from five paired forest–coffee-cleared sites (Table 1), because site differences can lead to large variation in atmospheric deposition rates.

We used one-way analysis of variance with post hoc multiple comparisons to test for differences in atmospheric deposition and throughfall ion fluxes among the three land-cover types. Independent-sample t tests were used to compare deposition rates and fluxes between seasons and years and to compare bulk and fog deposition samples within and between study regions. At each individual site, differences in net throughfall fluxes between forest and coffee land-cover types were examined with paired-sample t tests. Simple linear regression was used to examine relationships among anion fluxes, water inputs, and vegetation characteristics (e.g., leaf area index, LAI). Significance was set at $P < 0.05$ (two-tailed). Analyses were performed using SPSS version 16 (SPSS 2008).

Results

Bulk deposition and rainwater pH

In 2005–2006, bulk deposition to Xalapa-Coatepec was 8 ± 2 kg SO_4^{2-}-S/ha, 7 ± 1 kg Cl^-/ha, and 3 ± 0.3 kg NO_3^--N/ha (Fig. 2a). Deposition of all anions to XAC was nearly twofold greater (for S and N, $P < 0.04$) in 2006–2007 (14 ± 0.5 kg SO_4^{2-}-S/ha, 12 ± 1.5 kg Cl^-/ha, 5 ± 0.3 kg NO_3^--N/ha) than during the previous year. Sulfate-S and nitrate-N inputs were also significantly higher ($P < 0.05$, $P < 0.03$) than in Huatusco-Totutla, where bulk deposition in 2006–2007 was 8 ± 1 kg SO_4^{2-}-S/ha, 8 ± 2 kg Cl^-/ha, and 3.5 ± 0.1 kg NO_3^--N/ha. No differences were detected in bulk inputs among the collectors established in each region: SO_4^{2-}-S (XAC, $P < 0.43$; HUT, $P < 0.28$), Cl^- (XAC, $P < 0.78$; HUT, $P < 0.16$), and NO_3^--N (XAC, $P < 0.34$; HUT, $P < 0.87$). Two to three bulk collectors appeared sufficient to characterize XAC and HUT.

Our data did not reveal within-year seasonal differences in the magnitude of bulk deposition for any of the ions measured. Bulk deposition also displayed the same patterns during both sample years and across regions (Fig. 2a). Sulfate-S and nitrate-N inputs were slightly higher during wet season months, comprising 50–55% of annual bulk SO_4^{2-}-S and NO_3^--N deposition. Bulk Cl^- inputs were a little greater during the dry season and represented 56–60% of annual bulk chloride deposition.

Measurements of pH in bulk rainwater indicate that rainfall was acidic with a mean pH of 5.3 (range 4.5–6.1). Minimum values were recorded in December and maximum values in July. Significant seasonal differences in pH were detected, with rainwater more acidic during the dry (5.1) than during the wet season (5.6, $P < 0.008$). Rainwater became less acidic after passage through forest (5.4) and coffee (5.8) canopies, creating significant differences among the three cover types in dry ($F < 25.53$, $P < 0.00002$), but not wet season pH ($F < 3.37$, $P < 0.72$).

Fog deposition

Fog was an important deposition pathway during the dry season (Fig. 2b). In XAC, dry season fog deposition to passive collectors ranged from 2–3 kg SO_4^{2-}-S/ha, 3–7 kg Cl^-/ha, and 1.5–2 kg NO_3^--N/ha. In HUT, values ranged from 2–4 kg SO_4^{2-}-S/ha, 6–8 kg Cl^-/ha, and 1–3 kg NO_3^--N/ha. Deposition of all anions to fog collectors was greater during the dry than during the wet season. In XAC, these differences were significant for SO_4^{2-}-S ($P < 0.045$) and Cl^- ($P < 0.03$) in 2005–2006, but not in 2006–2007. In HUT, we also recorded a significant increase in SO_4^{2-}-S ($P < 0.045$), Cl^- ($P < 0.045$), and NO_3^--N ($P < 0.045$) fog inputs during the dry compared to the wet season. Fog chloride deposition displayed the most striking seasonal difference, with dry season fog Cl^- inputs as much as threefold greater than wet season Cl^- inputs.

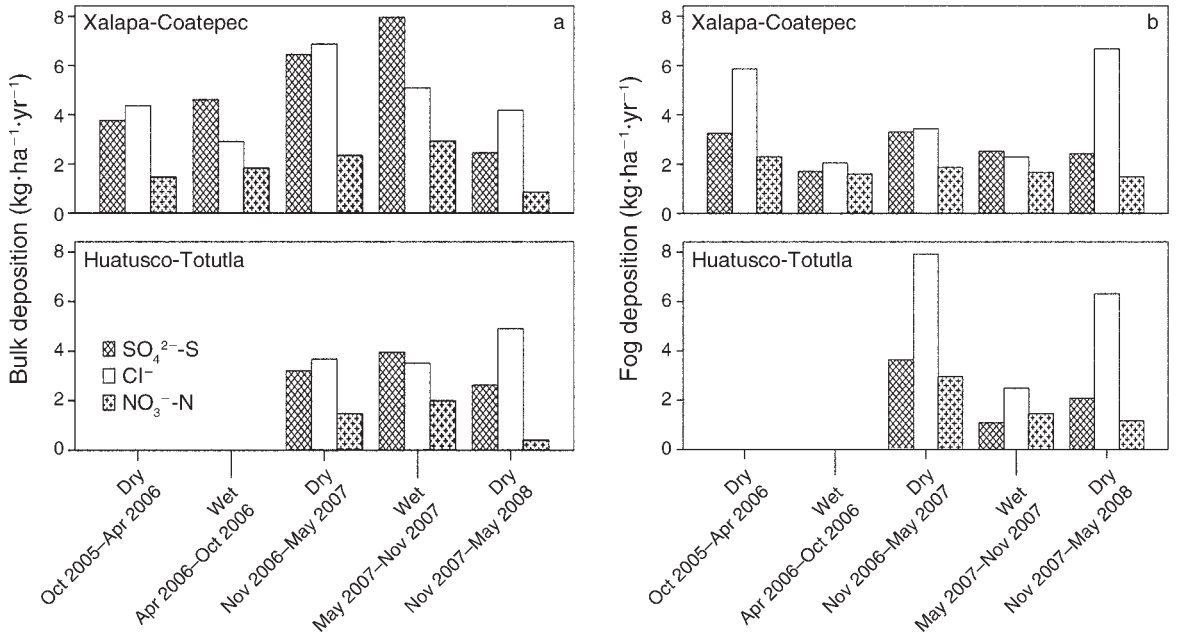

FIG. 2. (a) Bulk deposition and (b) fog deposition of SO_4^{2-}-S, Cl^-, and NO_3^--N to cleared areas in central Veracruz, Mexico. Between 2005 and 2008, deposition was measured for five consecutive seasons in Xalapa-Coatepec (XAC) and for three consecutive seasons in Huatusco-Totutla. Deposition data for the 2007–2008 dry season in XAC are for 23 January–5 April due to missing data for the 13 November–23 January sample period.

Total atmospheric deposition of SO_4^{2-}-S

We compared total atmospheric SO_4^{2-}-S deposition to forests, coffee agroforests, and clearings by calculating the mean sulfur flux per land-cover type for each region. We found differences between Xalapa-Coatepec and Huatusco-Totutla in both the magnitudes and spatial patterns of deposition. First, mean throughfall sulfur fluxes to forest and coffee soils were significantly greater in XAC than in HUT (forest, $F < 14.1$, $P < 0.003$; coffee, $F < 8$, $P < 0.015$; Fig. 3a). Second, SO_4^{2-}-S inputs to forests in XAC (17 ± 5 kg/ha, mean ± SE) were, on average, 41% and >100% higher than inputs to coffee agroforests (12 ± 1 kg/ha, $P < 0.15$) and cleared areas (8 ± 2 kg/ha, $P < 0.03$), respectively. By contrast, we did not detect differences in SO_4^{2-}-S input among forests (7 ± 1 kg/ha), coffee agroforests (7 ± 1 kg/ha),

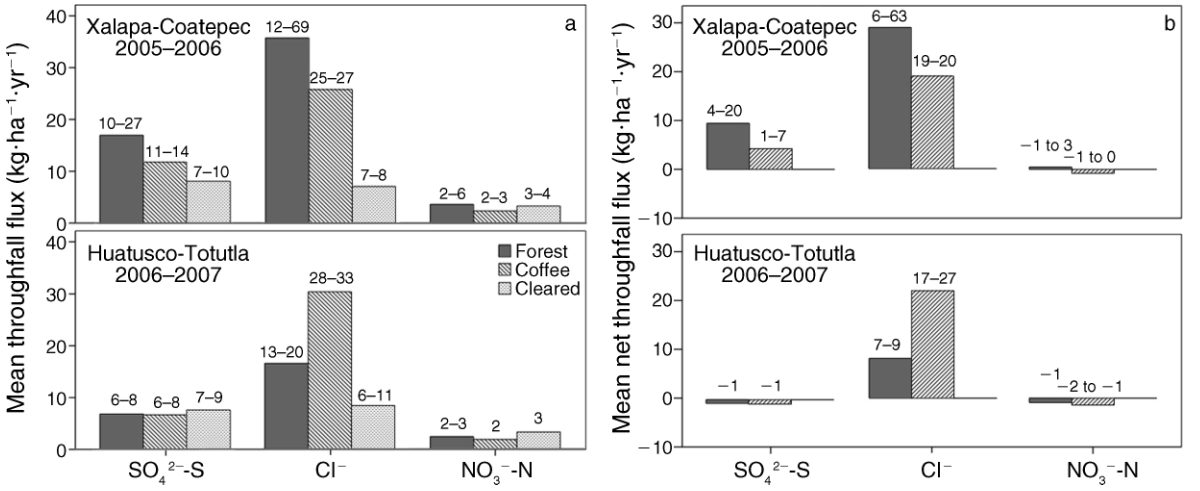

FIG. 3. (a) Mean throughfall flux and (b) mean net throughfall flux of SO_4^{2-}-S, Cl^-, and NO_3^--N to forest, shade coffee, and cleared land-cover types in central Veracruz, Mexico. Throughfall fluxes were measured in three forests and three coffee agroforests in Xalapa-Coatepec during 2005–2006, and in two forests and two coffee agroforests in Huatusco-Totutla from 2006 to 2007. Ranges of total and net throughfall ion fluxes to sites are indicated above the bars.

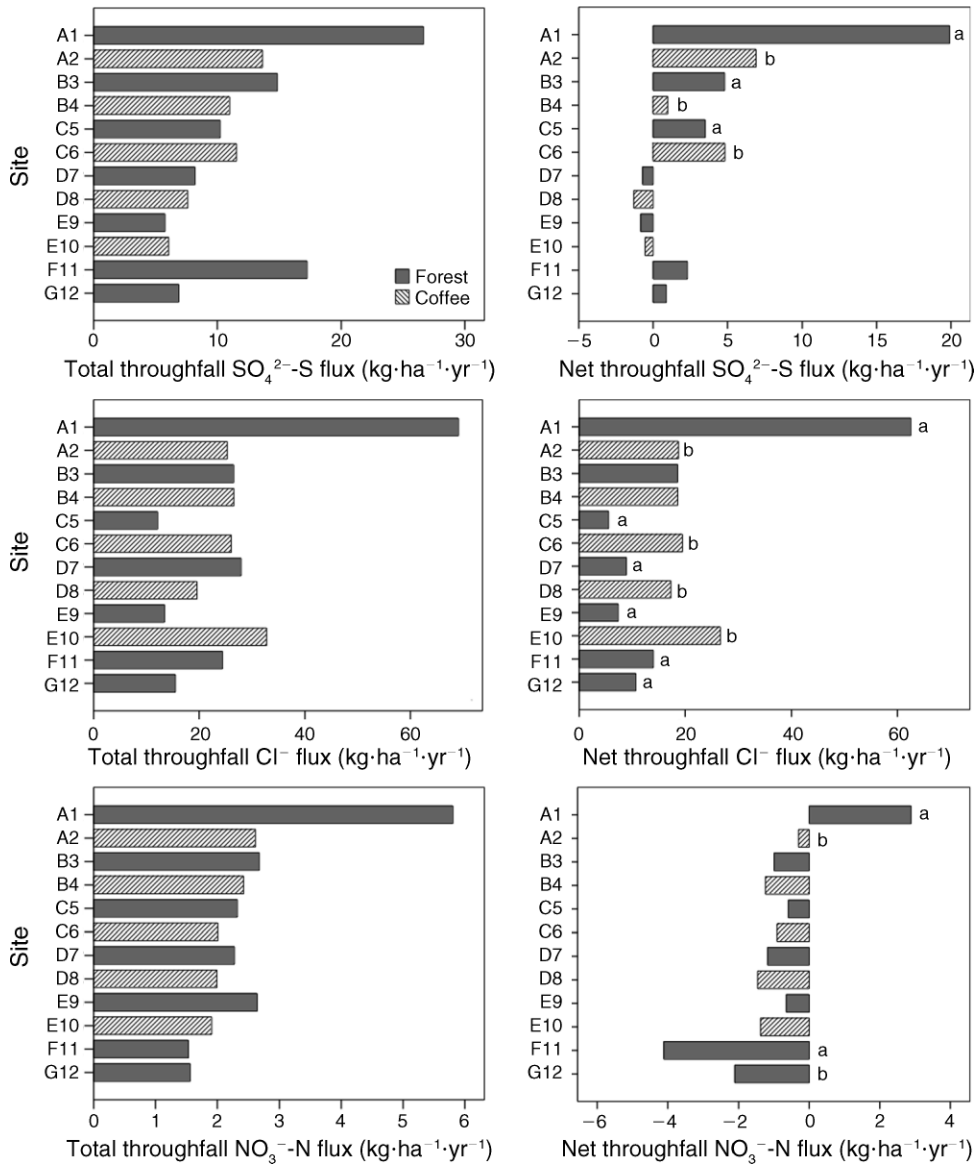

FIG. 4. Total throughfall flux and total net throughfall flux of sulfate-sulfur (SO_4^{2-}-S), chloride (Cl^-), and nitrate-nitrogen (NO_3^--N) to seven montane forests (gray bars) and five coffee agroforests (bars with diagonal lines) in central Veracruz, Mexico. Sites with the same capital letters indicate paired forest–coffee plots. Sites 11 and 12 are compared with cleared areas. Paired bars with different letters (a, b) indicate significant differences at the $P < 0.05$ level. For sites 11 and 12, "a" denotes a significant difference between forest and bulk deposition to adjacent clearing. Refer to Table 1 for site information.

and cleared areas (8 ± 1 kg/ha) in HUT ($P < 0.82$; Fig. 3a). Apart from these regional differences, greater variation was recorded in total S deposition within forest than coffee land cover. Throughfall SO_4^{2-}-S fluxes varied by more than fourfold across the seven forest, 6–27 kg/ha, and by twofold across the five coffee plots studied, 6–14 kg/ha (Fig. 4).

Overall, bulk sulfate-S deposition was much less variable than net (dry + fog) deposition (Fig. 3a, b). Indeed, net SO_4^{2-}-S fluxes to sites in XAC ranged from 4 to 20 kg/ha in forests and from 1 to 7 kg/ha in coffee agroforests (Fig. 4). Moreover, mean net SO_4^{2-}-S flux to forest (9 ± 5 kg/ha) was two times greater than to coffee land cover (4 ± 2 kg/ha; Fig. 3b), and this difference was significant ($P < 0.045$). In HUT, the net SO_4^{2-}-S input to each forest and coffee site was −1 kg/ha, equivalent to canopy uptake of 9–13% of bulk deposition. This apparent uptake of sulfur is small and represents an underestimate of total SO_4^{2-}-S deposition to forest and coffee cover types, because bulk samplers overestimate wet S deposition (thereby underestimating sulfur in net throughfall) and because not all SO_2 deposited through leaf stomata is released into throughfall (Lindberg and Garten 1988).

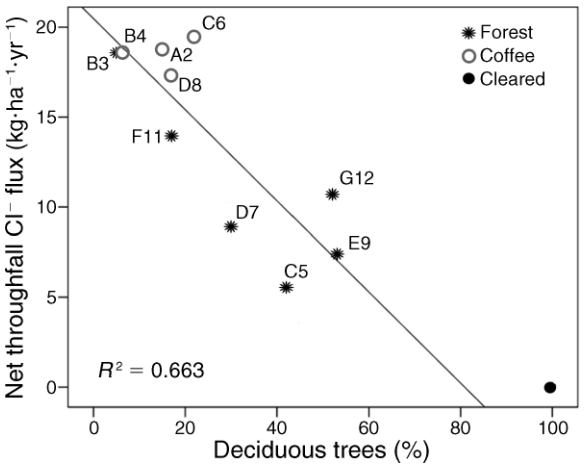

FIG. 5. Linear regression indicating a significant decrease in net annual throughfall Cl^- flux to forest fragments (stars) and coffee agroforests (open gray circles) in central Veracruz, Mexico, with increasing percentage of deciduous trees per plot. A cleared area with no canopy cover (i.e., 100% deciduous; solid circle) is inserted for reference.

We found a significant positive correlation between fog deposition and net SO_4^{2-}-S inputs to forest fragments in XAC ($r^2 = 0.834$, $P < 0.02$). This relationship did not hold for forest fragments in HUT or for coffee land cover in either study region. Although fogs occur during dry season months, we did not detect seasonal differences in net SO_4^{2-}-S inputs to either land-cover type (forest, $P < 0.65$; coffee, $P < 0.69$).

Chloride ion fluxes

Chloride in throughfall was at least twofold greater than sulfate-S in throughfall (Fig. 3a). The range of spatial variation in total Cl^- fluxes was also wider than for sulfate-S or nitrate-N. Between 12 and 69 kg Cl^-/ha were delivered to forest and between 25 and 33 kg Cl^-/ha to coffee land cover (Fig. 4). Although we found significant differences among forest, coffee, and cleared land-cover types in Xalapa-Coatepec ($F < 7.79$, $P < 0.04$) and in Huatusco-Totutla ($F < 11.69$, $P < 0.01$), spatial patterns of total throughfall Cl^- inputs differed between the regions sampled (Fig. 3a). In XAC, forest plots received 36 ± 17 kg Cl^-/ha (mean ± SE), while coffee agroforests received 26 ± 0.3 kg Cl^-/ha, but these differences were not significant ($P < 0.53$). Even with the exclusion of A1 (a statistical outlier), average Cl^- flux to forest (19 ± 7 kg/ha) was not significantly different ($P < 0.13$) from average Cl^- flux to coffee (26 kg ± 0.4 kg/ha). This pattern was reversed in HUT, where mean Cl^- input to coffee (30 ± 2 kg/ha) was significantly higher ($P < 0.02$) than to forest (17 ± 3 kg/ha).

The amount of Cl^- delivered in throughfall was consistently higher than the amount deposited to clearings. Depending on the site, there was a two- to 10-fold enrichment of throughfall with Cl^- relative to bulk deposition (Fig. 4). On average, NTF chloride inputs to coffee were significantly greater than to forest sites in XAC (coffee, 19 ± 0.2 kg/ha; forest without A1, 12 ± 6.5 kg/ha; $F < 9.31$, $P < 0.01$) and in HUT (coffee, 22 ± 5 kg/ha; forest, 8 ± 1 kg/ha; $F < 11$, $P < 0.006$; Fig. 3b). These forest–coffee differences in NTF were significant during the dry (XAC, $P < 0.05$; HUT, $P < 0.005$), but not during the wet season (XAC, $P < 0.14$; HUT, $P < 0.1$).

Cl^- in net throughfall was strongly related to canopy composition. We found that net Cl^- fluxes (outlier removed) decreased significantly with increasing abundance of deciduous trees (NTF Cl^- flux $= 20.48 - 0.25 \times$ percentage deciduous trees, $R^2 = 0.66$, $P < 0.03$; Fig. 5). In forest sites, net Cl^- fluxes were also significantly and positively correlated with throughfall water inputs, but only when site A1 was excluded from analysis ($r^2 = 0.64$, $P < 0.02$; $r^2 = -0.03$, $P < 0.92$, outlier included). In coffee sites, net Cl^- inputs were positively related to throughfall water fluxes as well, but the correlation ($r^2 = 0.48$) was significant at the $P < 0.1$ level. These correlations point to increased rates of foliar chloride leaching during high rainfall periods.

Nitrate-N ion fluxes

Relative to sulfate-S and chloride, nitrate-N fluxes were low and displayed the least variation among land-cover types (Fig. 3a). In XAC and HUT, mean nitrate-N input to forest (XAC, 3.6 ± 1 kg/ha; HUT, 2.5 ± 0.2 kg/ha), shade coffee (XAC, 2 ± 0.2 kg/ha; HUT, 2 ± 0.4 kg/ha) and cleared land-cover types (XAC, 3 ± 0.4 kg/ha; HUT, 3 ± 0.1 kg/ha) was similar ($F < 0.92$, $P < 0.412$). Moreover, there was little recorded variation in total NO_3^--N flux among sites (2–6 kg/ha; Fig. 4).

Net NO_3^--N fluxes were negative at all but one site (Fig. 4). Net uptake of NO_3^--N varied much more widely than total fluxes, from −1 to −4 kg NO_3^--N/ha in forest and from −0.3 to −1.5 kg NO_3^--N/ha in coffee sites. At only one forest site were NO_3^--N inputs positive. We did not find clear relationships between net nitrate-N fluxes and water inputs or vegetation variables.

Deposition hotspot

Total and net sulfate, chloride, and nitrate fluxes were comparatively high at site A1. Net SO_4^{2-}-S and Cl^- inputs to this site were as much as sixfold higher while NO_3^--N inputs were fourfold higher than measured inputs to all other sites (Fig. 4). Net fluxes were positively correlated with fog deposition of sulfate-S ($r^2 = 0.76$, $P < 0.05$), Cl^- ($r^2 = 0.77$, $P < 0.45$), and nitrate-N ($r^2 = 0.82$, $P < 0.02$).

DISCUSSION

Our results confirm that land cover significantly influences atmospheric inputs to ecosystems. However, there was remarkable variability in the magnitudes and spatial patterns of ion flux across this tropical montane

landscape. Below, we discuss possible mechanisms influencing these patterns as well as the management and ecological implications of our findings.

Controls on atmospheric deposition and throughfall fluxes

Sulfur in throughfall.—We expected that net SO_4^{2-}-S inputs would increase with canopy leaf area (until a saturation point where forest canopies become too dense to efficiently filter particles; e.g., Lovett and Reiners 1986) due to a previously demonstrated positive relationship between fog and dry deposition and collection surface area (Draaijers et al. 1992). Contrary to expectation, this pattern was not observed in one of the study regions (i.e., HUT; Fig. 3a). Three possible and potentially interconnected explanations include: (1) local differences in air quality; (2) intra- and inter-site differences in canopy structure, topographic exposure, and elevation; and (3) effects of interannual precipitation variability on atmospheric deposition. We did not sample throughfall chemistry in the same location for two consecutive years. Therefore, reasons for this divergence remain unclear.

Long-term monitoring of air concentrations in the United States shows that the relative contribution of dry SO_2 and aerosol SO_4^{2-} deposition to ecosystems is strongly related to distance from local and regional emission sources (CASTNET 2007 annual report; *available online*).[5] In 2007, dry deposition measured at CASTNET sites comprised 7–54% of total S deposition, with lower inputs at western coastal sites and higher inputs near industrial sources in the Midwest (see footnote 5). The Xalapa-Coatepec and Huatusco-Totutla regions are 35 km apart and similarly distant from the Gulf of Mexico and industrial emission sources along the coast. The similar spatial location of XAC and HUT relative to these sources suggests that higher rates of fog and dry deposition to, and land-cover differences expressed in, XAC could be due to higher atmospheric sulfur concentrations there. In the Mexico City Basin, Pérez-Suárez et al. (2008) measured throughfall sulfate-S fluxes under fir and pine forest at sites ~40 km apart with high and low deposition. Comparable to our results, they found that net SO_4^{2-}-S fluxes were three- to fivefold greater at the high than at the low deposition site and that dissimilarities between fir and pine were magnified at the more polluted site. We do not have sufficient data to ascertain the magnitude of local influences (i.e., $\leq$20 km radius) on the atmospheric environment in central Veracruz. Yet, unlike Huatusco-Totutla, two diesel-burning sugar mills (Fig. 1), a major dairy processing factory, and lime manufacturing operations operate within the vicinity of Xalapa-Coatepec. Therefore, it is a reasonable hypothesis that sulfur emissions generated by these industries during fuel combustion could contribute to increased pollutant loading in the Xalapa-Coatepec region. However, no emissions data are currently available to substantiate this supposition.

Site-level differences in fog and dry SO_4^{2-}-S inputs also obscured the effects of land-cover type on total atmospheric sulfur deposition (Fig. 4). While we held slope and aspect constant in our experimental design, the fragmented nature of this landscape prevented us from simultaneously controlling for canopy characteristics and topographic exposure. Across the forest sites, basal area and tree density ($\geq$5 cm diameter at breast height) varied more than two- and fourfold, respectively, and similarity in tree species composition was low (Ponette-González et al. 2010). In addition, sampling spanned a 1200-m altitudinal gradient. These factors are known to strongly affect fog and dry deposition rates (Lovett and Reiners 1986, Weathers et al. 1988, 1992, 2000, Lovett et al. 1999), and, combined, certainly contributed to the heterogeneous patterns of sulfur fluxes reported here.

Our findings suggest that interannual variation in meteorological conditions was another important mechanism driving variation in S fluxes (Kelly et al. 2009). The amount and timing of dry season rainfall displayed a striking difference between water years 2005–2006 and 2006–2007. In 2006–2007, a winter El Niño event was associated with unusually high precipitation during peak dry months (Ponette-González et al. 2010). Drier conditions in 2005–2006 would be expected to result in enhanced dry SO_4^{2-}-S inputs to ecosystems with greater canopy cover. Conversely, wetter conditions and higher wet deposition rates in 2006–2007 could explain why land-cover differences were not evident in Huatusco-Totutla. Site comparisons show that net SO_4^{2-}-S inputs to sites in XAC were substantial in 2005–2006 (sites A–C; Fig. 4), while low inputs to forests in XAC were measured in 2006–2007 (sites F–G; Fig. 4) when bulk S deposition was twofold higher than in 2005–2006 (Fig. 2a).

Chloride.—The shift in species composition from deciduous to evergreen-dominant associated with forest to coffee land-cover change increased net chloride inputs to coffee soils by a factor of two to four (Fig. 5). We suggest that some combination of enhanced fog capture, dry deposition, and/or foliar leaching by coffee canopies during dry season months contributed to the significant increase in net Cl^- flux to coffee agroforests. Two lines of evidence support this view. First, dry season fog water inputs to shade coffee soils were consistently higher than inputs to neighboring forests due to lower rates of canopy water retention and/or evaporation (Ponette-González et al. 2010). Although this appears surprising, canopy thinning can result in increased or decreased fog water fluxes, depending upon LAI (leaf area index; Lovett and Reiners 1986, Ataroff and Rada 2000). Furthermore, our recorded spike in dry season chloride deposition to fog samplers (Fig. 2b) is indication that fog events were chemically concentrated with Cl^- (e.g.,

[5] ⟨http://www.epa.gov/castnet/index.html⟩

TABLE 2. Bulk deposition, throughfall, and net throughfall fluxes in $kg \cdot ha^{-1} \cdot yr^{-1}$ sampled under tropical forests in Latin America and eastern temperate forests in the United States.

Site	Elevation (m a.s.l.)	Rainfall (mm)	Sea (km)	Bulk deposition			Total throughfall flux		
				SO_4^{2-}-S	Cl^-	NO_3^--N	SO_4^{2-}-S	Cl^-	NO_3^--N
Tropical broadleaf									
Mexico†	1100	1339	83	8.9	10.7	3.5	8.2	19.6	2.3
Mexico†	1350	1788	92	6.6	6.1	3.3	5.8	13.4	2.6
Mexico†	1350	2376	72	10.0	8.0	3.7	14.8	26.6	2.7
Mexico†	1450	1873	67	6.7‡	6.6‡	2.9‡	26.6	69.1	5.8
Mexico†	1500	2001	68	6.7‡	6.6‡	2.9‡	10.2	12.1	2.3
Mexico†	1600	2421	71	14.9	10.4	5.6	17.2	24.4	1.5
Mexico†	2200	1105	70	6.0	4.8	3.7	6.9	15.5	1.6
Mexico (mean)	1100–2200	1843	75	9	8	4	13	26	3
Panama	1200	3678	<30	13.0	34.5	7.0	6.0	50.0	7.0
Puerto Rico	265–456	3482	10	46.0	140.9	1.5	54.4	120.8	2.2
Ecuador	1900–2200	2220	>200	1.1	12.0	3.0	4.0	24.0	8.0
Brazil	1050	1951	15	3.2	10.6	2.4	10.1	62.5	2.1
Brazil	798	1453	50	10.8	12.9	13.9	19.6	26.0	10.8
Colombia	2550	2115	~200	26.2	19.4	ND	40.9	36.3	ND
Colombia	3770	1453	~200	16.8	13.6	ND	31.7	19.8	ND
Jamaica	1809	3060	<30	<4.1	34.9	<1.7	9.9	42.7	<0.5
Jamaica	1824	3060	<30	<4.1	34.9	<1.7	<6.9	29.6	<0.5
Mexico City	~3300	778–1455					9–20		3–11
Temperate									
Acadia†									
Conifer	7–413	1286	2	4.8	5.1	2.6	10.5	25.2	5.3
Deciduous	7–413	1286	2	4.8	5.1	2.6	6.6	11.3	2.9
Forest (mean)	7–413	1286	2	4.8	5.1	2.6	9.0	19.8	4.3
Catskills†									
Conifer	524–1265	1650	140	9.4	2.4	1.6	12.2	5.9	1.8
Deciduous	524–1265	1650	140	9.4	2.4	1.6	10.1	4.3	2.1
Forest (mean)	524–1265	1650	140	9.4	2.4	1.6	11.3	5.1	2.2
GRSM†									
Conifer	579–2012	1439	450	7.2	2.3	2.3	38.5	12.2	5.7
Deciduous	579–2012	1439	450	7.2	2.3	2.3	6.5	2.7	1.2
Forest (mean)	579–2012	1439	450	7.2	2.3	2.3	28.5	9.2	4.9

Note: Sampling was conducted for a minimum of one year at all tropical sites. "ND" indicates no data.
† Anion-exchange resin throughfall sampling employed as method of collection.
‡ Throughfall fluxes to these sites were compared to the same bulk deposition collector.
§ Unpolluted watershed.
¶ Polluted watershed.

Weathers et al. 1988). Second, while our findings reveal the possibility of enhanced dry deposition and/or foliar Cl^- leaching from forest and coffee shade trees, and potentially coffee leaves as well (Tukey 1970), in this region, forest LAI may decrease by 40% during dry season months. In contrast, LAI in evergreen shade coffee stands does not vary seasonally (Ponette-González et al. 2010). Previous studies show that reductions in foliar Cl^- leaching from forest canopies during leaf-off periods can reduce Cl^- levels in forest canopy throughfall (Potter et al. 1991, Berger et al. 2001).

These findings combined may also explain the wide range in Cl^- fluxes we observed, and why forest fragments displayed greater spatial variation in net Cl^- fluxes than coffee sites (Fig. 4). In deciduous forests, complete leafless periods decrease throughfall spatial variability (Staelens et al. 2006). In highland Veracruz, asynchronous leaf abscission by forest trees (Williams-Linera 1997) coupled with the structural variability of forest stands appears to amplify spatial variation in throughfall ion fluxes.

Nitrate-N.—We documented canopy nitrate-N retention at all, but one site, where nitrate inputs to the forest floor doubled as a result of net N leaching. Forest rates of uptake varied from 20% to 73% of bulk NO_3^--N deposition, comparable to values reported from Jamaican montane rainforests (Hafkenscheid 2000; Table 2). Coffee canopies absorbed similar amounts (10–42%) of nitrate-N as both forests and shade coffee plantations in Colombia (Jaramillo-Robledo 2003). In this latter study, 29–60% of NO_3^--N in precipitation was retained in coffee plots shaded with *Inga*, *Pinus*, and *Eucalyptus*; only in a *Cordia*-shaded plot was net leaching of NO_3^--N recorded.

The role of trees and epiphytes in altering throughfall nitrogen fluxes has been widely reported in tropical forests (Lang et al. 1976, Clark et al. 1998*b*, Hölscher et al. 2003). In Monteverde, Costa Rica, a montane cloud forest canopy retained 80% of total atmospheric nitrate-

TABLE 2. Extended.

Net throughfall flux			
SO_4^{2-}-S	Cl^-	NO_3^--N	Source
−0.7	8.9	−1.2	see *Results*
−0.8	7.4	−0.7	see *Results*
4.8	18.6	−1.0	see *Results*
19.9	62.5	2.9	see *Results*
3.5	5.5	−0.6	see *Results*
2.3	14.0	−4.1	see *Results*
0.9	10.7	−2.1	see *Results*
4	18	−1	
−7.0	15.0	−0.1	Cavelier et al. (1997)
8.4	−20.1	0.7	Heartsill-Scalley et al. (2007)
2.9	12.0	5.0	Wilcke et al. (2001)
6.9	51.9	−0.4	Forti et al. (2007) §
8.8	13.1	−3.1	Forti et al. (2007) ¶
14.7	17.0	ND	Veneklaas (1990)
14.9	6.2	ND	Veneklaas (1990)
>5.8	7.8	~−1.2	Hafkenscheid (2000)
~2.8	−5.3	~−1.2	Hafkenscheid (2000)
			Fenn et al. (1999)
			Weathers et al. (*unpublished data*)
5.7	20.1	2.7	
1.8	6.2	0.3	
4.2	14.7	1.7	
			Weathers et al. (*unpublished data*)
2.8	3.5	0.2	
0.7	1.9	0.5	
1.9	2.7	0.6	
			Weathers et al. (*unpublished data*)
31.3	9.9	3.4	
−0.7	0.4	−1.1	
21.3	6.9	2.6	

N deposition (Clark et al. 1998*b*). Another study by Clark et al. (2005) reveals the key role of epiphytic bryophytes in the uptake of atmospherically deposited nitrogen. Epiphytic organisms can also contribute to a net addition of nitrogen to the forest floor. In a comparison of temperate and tropical rain forests, Nadkarni (1986) found that N leaching by epiphytes was greater than N accumulation during wet season months. These and other studies highlight the potential for great site-level variability in canopy N cycling due to variations in environmental conditions (e.g., rainfall seasonality), epiphyte functional groups, and tree species composition.

Notably, percentage canopy NO_3^--N uptake is often calculated as the net difference between throughfall flux and bulk, rather than total, NO_3^--N deposition. Thus, many of these studies (including ours) overestimate nitrate-N uptake by vegetation canopies, because dry and fog N deposition can be substantial. For example, based on three years of measurements, Galy-Lacaux et al. (2003) concluded that deposition of gaseous N was 75% of total deposition to tropical rain forest in west central Africa. In northern Peru, modeled dry N inputs to tropical cloud forest were 2–24 $kg \cdot ha^{-1} \cdot yr^{-1}$ compared to measured wet inputs of 8–13 $kg \cdot ha^{-1} \cdot yr^{-1}$ (Wilcke et al. 2008). Estimated N inputs from fog are also high, 13–35% in the San Bernardino Mountains (Fenn et al. 2000) to 96% of total N deposition on the island of Hawai'i (Heath and Huebert 1999).

Data from our passive fog samplers show that fog may serve as an important vector of NO_3^--N deposition to ecosystems in eastern Mexico as well. Fog water inputs comprised ~16% of total above-canopy dry season precipitation (Ponette-Gonzalez et al. 2010), yet contributed 66% of bulk plus fog nitrate-N deposition during the dry season.

A regional perspective

The magnitudes of anion fluxes in the central Veracruz highlands are similar to those measured in some polluted tropical and temperate montane regions. Atmospheric SO_4^{2-}-S deposition to forests and coffee agroforests was intermediate between tropical sites influenced by volcanic SO_2 emissions (e.g., Veneklaas 1990, Heartsill-Scalley et al. 2007) and sites distant from sea and anthropogenic sources (Wilcke et al. 2001; Table 2). On average, total annual sulfur flux to forested sites was comparable to values reported for forests in the Mexico City Basin that receive 9–20 kg S/ha (Fenn et al. 1999). This finding surprised us not only because Mexico City is one of the most polluted metropolitan areas in the world, but also because Fenn et al. (1999) suggest that emissions from nearby Popocatépetl volcano increased deposition rates during their study. Furthermore, in these studies, deposition estimates were made for conifer stands that incur substantially higher inputs than broadleaf forests (Weathers et al. 2006*a*).

Although chloride represented the largest anion flux that we measured to forest and coffee soils, inputs fell at the low end of the range when compared with Neotropical forests. Values in Table 2 suggest that this was largely the combined result of lower annual rainfall and decreased seasalt influence on precipitation chemistry in our sampling locations. Conversely, Cl^- fluxes to our sites were much higher than to forests in an inland mountain range in the eastern United States (Catskill Mountains; Table 2), where HCl emissions from fossil-fuel combustion (although now decreasing) are a known source of chloride in atmospheric deposition (Lovett et al. 2005).

Nitrate-N in throughfall collected in high-elevation forests downwind of polluted urban areas often exceeds 5 $kg \cdot ha^{-1} \cdot yr^{-1}$ (Fenn et al. 1999, Weathers et al. 2006*a*). Compared with these sites, throughfall NO_3^--N inputs measured in our study were relatively low. Nonetheless, total NO_3^--N deposition to forest and coffee ecosystems in central Veracruz is likely to be much greater due to canopy interactions not accounted for with throughfall measurements (Fenn et al. 2000).

To estimate total NO_3^--N deposition to plant canopies, we calculated bulk : fog + dry ratios using net sulfur as an index of fog plus dry deposition. On

PLATE 1. In recent decades, low coffee prices have stimulated conversion of shade coffee agroforests, such as this one near the city of Coatepec, Veracruz State, Mexico, for residential development. Photo credit: A. G. Ponette-González (March 2006).

average, fog plus dry sulfur inputs comprise 33% of total S deposition to forest and 25% of total deposition to coffee canopies. Assuming bulk : fog + dry ratios of 3:1 (forest) and 4:1 (coffee), total annual NO_3^--N deposition is 4–7 $kg \cdot ha^{-1} \cdot yr^{-1}$ and 3.5–4.4 $kg \cdot ha^{-1} \cdot yr^{-1}$, respectively. These rates fall below the critical threshold of 10–15 $kg \cdot ha^{-1} \cdot yr^{-1}$ established for temperate forests (Achermann and Bobbink 2003), the limit below which ecosystems are not expected to experience negative effects from pollutant exposure. However, we did not estimate ammonium fluxes, which may be of equivalent magnitude (Báez et al. 1997).

Annual bulk S and N fluxes were nine- and eightfold greater, respectively, than background wet deposition rates for remote tropical areas (1–3 kg S/ha [Rodhe et al. 1995]; 0.5 kg N/ha [Galloway et al. 2008]). Comparisons with comparable sites using long-term data from the U.S. National Atmospheric Deposition Monitoring Program also indicate relatively elevated deposition in our study region (NADP 2009). Bulk Cl^- inputs to highland Veracruz were within the range of levels reported for U.S. sites receiving the highest Cl^- deposition (4.5–23 $kg \cdot ha^{-1} \cdot yr^{-1}$; NADP 2009), but bulk SO_4^{2-} (28 kg SO_4^{2-}/ha) and bulk NO_3^- deposition (17 kg NO_3^-/ha) were similar to wet deposition rates in the most polluted locales in the northeastern United States (>27 kg $SO_4^{2-} \cdot ha^{-1} \cdot yr^{-1}$, >20 kg $NO_3^- \cdot ha^{-1} \cdot yr^{-1}$). In addition, during the span of our measurements, annual bulk deposition to Xalapa was substantially higher than measured in the mid-1990s (Báez et al. 1997; Table 3). Although this difference may partially reflect contrasts in sampling methodology (i.e., wet-only vs. bulk sampling), studies clearly show that since the 1990s air quality has continued to decline in the region of Mexico in which we sampled (Parungo et al. 1990, Bravo et al. 2006).

Sources in the Gulf of Mexico watershed.—Diverse natural and anthropogenic emission sources have been shown to contribute S, Cl^-, and N in the Gulf of Mexico watershed (Parungo et al. 1990, NEI 2006, Millet et al. 2009). A few of these anthropogenic sources are likely to increase future deposition to montane ecosystems in the Sierra Madre Oriental. According to Mexico's National Emissions Inventory (NEI 2006), electricity generation and oil and gas extraction with refining are the largest point sources of SO_x and NO_x along Mexico's Gulf coastline. In 1999, the State of Veracruz ranked fourth nationally in SO_x emissions (~342 000 Mg) after Puebla and Jalisco, where degassing volcanoes are the primary polluters, and second in NO_x emissions (~162 000 Mg) after Coahuila (NEI 2006). Located in the City of Tuxpan, the Adolfo López Mateos power plant alone produced ~75% and ~10% of these totals, respectively (Miller and Van Atten 2004). In addition to power plants, Mexico's petrochemical industry is concentrated in the Gulf coast region, primarily in Tabasco, SE Veracruz, and Tamaulipas. These industries require

TABLE 3. Bulk deposition and rainfall measured in Xalapa, Mexico, from 1993 to 1995 and from October 2005 to April 2008.

	Wet season (kg/ha)			Dry season (kg/ha)			Annual (kg/ha)			
Year	SO_4^{2-}-S	Cl^-	NO_3^--N	SO_4^{2-}-S	Cl^-	NO_3^--N	SO_4^{2-}-S	Cl^-	NO_3^--N	Rainfall (mm)
1993†	0.9	0.6	0.2	ND	ND	ND	ND	ND	ND	1513
1994†	2.8	1.3	1.1	2.3	0.6	0.5	5.1	1.9	1.5	1187
1995†	3.2	1.5	1.1	1.9	0.6	0.4	5.1	2.1	1.5	1448
2005	3.6	2.3	1.6	3.2	4.3	1.3	6.7	6.6	2.9	1451
2006	8.8	5.3	3.0	5.1	8.2	1.8	13.9	13.5	4.9	1907
2007	ND	ND	ND	3.6	5.8	1.1	ND	ND	ND	1443

† Data for 1993–1995 are from Báez et al. (1997). Deposition data for the 2007 dry season are for 23 January–5 April due to missing data for the 13 November–23 January sample period.

chlorine as essential feedstock for chemical manufacturing (e.g., vinyl chloride) and emit significant amounts of chlorinated (Millet et al. 2009) and sulfur-containing compounds that can be incorporated into rain and fog and redeposited to downwind ecosystems (Báez et al. 1997, Raja et al. 2005). Growing demand for electricity (Cancino-Solórzano et al. 2010) and reactivation of Mexico's petrochemical sector (Laguna 2004) suggest that atmospheric inputs from these regional sources will continue to increase in the near future.

N deposition is also likely to increase near rapidly urbanizing areas (e.g., Xalapa) from vehicle NO_x emissions and increasing demand for sugarcane-based ethanol (Jank et al. 2007). Sugarcane burning is a well-documented source of NO_x in the tropics (Crutzen and Andreae 1990) and has been found to elevate N deposition in adjacent montane areas (Boy et al. 2008).

Management and ecological implications

Hotspots of deposition as a result of spatial heterogeneity.—While specific regions in the Sierra Madre Oriental may be experiencing elevated deposition, our findings highlight the differential vulnerability of montane sites to air pollution. For example, we measured extraordinarily high anion fluxes to a small (~3 ha) hilltop forest patch (A1; Fig. 4). Such hilltops have been shown to receive higher deposition from increased wind and fog exposure (Delmelle et al. 2002, Weathers et al. 2006*a*), thus we expected ion fluxes and fog deposition to be correlated at this site. However, this site was also located directly upslope from cattle pasture. Deposition to the upwind forest edge (where the collectors were located) was likely enhanced due to increased fog and aerosol impaction rates that resulted from upslope air flow (Fowler et al. 1989, Asbury et al. 1994) and contrasting vegetation structure between forest and grassland (Weathers et al. 2000, Ewing et al. 2009). In addition, this site's oak-dominated canopy is covered with *Tillandsia usneoides* L. (Spanish moss, Bromeliaceae). *Tillandsia usneoides* is an efficient scavenger of atmospheric nutrients and pollutants and has been shown to enhance throughfall ion concentrations below oak canopies (Schlesinger and Marks 1977).

These findings illustrate how topography, vegetation structure, and spatial arrangement of land-use systems combine in distinctive ways to influence nutrient and pollutant fluxes across human-modified montane landscapes. From a management perspective, this spatial heterogeneity in deposition indicates that there may be identifiable hotspots of species and ecosystems at risk of excess and increasing deposition. For example, in the western region of Xalapa, ~30% of forest patches occur on east-facing (in the direction of prevailing winds) hillslopes, while ~59% are adjacent to cattle pastures (Williams-Linera et al. 2002). Our results demonstrate that these forests are likely to receive elevated deposition loads. Future monitoring activities should encompass such hotspots.

Ecological effects.—Our study shows that elevated deposition may represent an additional environmental stress for Mexico's highly diverse tropical montane forests, which currently occupy <1% of the land area yet harbor ~12% of the nation's plant species (Rzedowski 1996). Coupled with the well-documented effects of fragmentation and anthropogenic disturbance on tree populations (Williams-Linera 2002, Ramírez-Marcial 2003), increased rates of nutrient and pollutant deposition may directly affect forest species composition and regeneration dynamics through such mechanisms as phytotoxicity (Alvarado-Rosales and Hernández-Tejeda 2002). More indirectly, pollution may alter species' ability to acquire resources (DeHayes et al. 1999). In Mexico City's urban forests, direct (i.e., ozone injury) and indirect (i.e., nutritional deficiencies) factors have been evoked to explain the decline of sacred fir (*Abies religiosa*), a common species throughout the mountains of central and southern Mexico and Guatemala.

Chronic inputs of sulfate, nitrate, and chloride have also been shown to induce base cation leaching and soil acidification in several ecosystem types in eastern North America (Weathers and Lovett 1998, Driscoll et al. 2001, Lovett et al. 2009). In this study, four sites fell within the "high" (i.e., high soil buffering capacity), while six sites were close or exceeded the "low" critical load ranges for sulfur proposed by Kuylenstierna et al. (2001). This suggests that certain sites in central Veracruz are at risk of acidification. Nevertheless, anion deposition effects on these tropical Andosols will depend on a host of biological, soil, and geological factors (Lovett et al. 2009), including N limitation and biotic N uptake (Matson et al. 1999, Fenn et al. 2002), sulfate-adsorption capacity (Pérez-Suárez et al. 2008), chloride and nitrate retention (Delmelle et al. 2003, Lohse and Matson 2005), and weathering rates (Vitousek 2004).

We conclude that land-cover change, climate change, and increasing atmospheric deposition are likely to affect tropical montane ecosystems through alterations in biogeochemical cycles. Yet, critically important gaps remain in our knowledge of deposition rates and patterns across most of the tropics. In highland areas, these data are urgently needed to assess potential sources and strength of off-site emissions as well as effects on species diversity and ecosystem function.

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