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Complex topography influences atmospheric nitrate deposition in a neotropical mountain rainforest

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HIGHLIGHTS

- Long term measurements of nitrate inputs through occult precipitation and rain along an altitudinal gradient.
- The transport of nitrate precursors was modeled based on emission inventories and satellite data.
- Transported NO_x was only correlated to nitrate concentrations in occult precipitation water from well exposed terrain peaks.
- Biomass burning NO_x was the most probable source explaining the variability of nitrate depositions.

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ABSTRACT

Future increase of atmospheric nitrogen deposition in tropical regions is expected to have negative impacts on forests ecosystems and related biogeochemical processes. In tropical mountain forests topography causes complex streamflow and rainfall patterns, governing the atmospheric transport of pollutants and the intensity and spatial variability of deposition. The main goal of the current study is to link spatio-temporal patterns of upwind nitrogen emissions and nitrate deposition in the San Francisco Valley (eastern Andes of southern Ecuador) at different altitudinal levels. The work is based on Scanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) retrieved-NO_2 concentrations, NO_x biomass burning emissions from the Global Fire Emissions Database (GFEDv3), and regional vehicle emissions inventory (SA-INV) for urban emissions in South America. The emission data is used as input for lagrangian atmospheric backward trajectory modeling (FLEXTRA) to model the transport to the study area. The results show that NO_3^- concentrations in occult precipitation samples are significantly correlated to long-range atmospheric secondary nitrogen transport at the highest meteorological stations (MSs) only, whereas for NO_3^- concentrations in rain samples this correlation is more pronounced at the lower MSs. We conclude that ion concentrations in occult precipitation at the uppermost MSs are mainly linked to distant emission sources via the synoptic circulation impinging the more exposed higher sites. Lower correlations close to the valley bottom are due to a lower occult precipitation frequency and point to a contamination of the samples by local pollution sources not captured by the used emission data sources.

1. Introduction

Nitrogen oxide emissions (NO_x) are increasing at a global scale because of population growth, increasing industrial activity, and greater land use change rates (Lewis et al., 2004). This also holds true for nitrate and other nitrogen species (Matson et al., 1999, 2002; Galloway et al., 2004). The unfavorable development especially applies to developing countries like the growing economies of South America, which are rapidly becoming the major nitrogen emitters of the world prone to enhanced atmospheric deposition (Vallack et al., 2001). For many of valuable ecoregions, N

List of abbreviations: RBSF, Reserva Biológica San Francisco; SCIAMACHY, Scanning Imaging Absorption SpectroMeter for Atmospheric CHartographY; EDGAR, Emission Database for Global Atmospheric Research; RETRO, REanalysis of the TROpospheric chemical composition over the past 40 years; GFEDv3, Fire Emission Database version 3; SA-INV, regional vehicle emissions inventory; MS, Meteorological Station; FLEXTRA, lagrangian atmospheric trajectory model; Nr, Reactive nitrogen; ITCZ, Inter Tropical Convergence Zone; CASA-GFED, Carnegie Ames Stanford Approach-Global Fire Emission Database.

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accumulation is probably the main driver of changes to species composition by changing competitive interactions in the ecosystems. Bobbink et al. (2010) showed that particularly tropical parts of Latin America as the Andes of Ecuador, hitherto not receiving enough attention, belong to the vulnerable regions in the next decades.

The effects of Nr (Reactive nitrogen) deposition in temperate terrestrial ecosystems related to anthropogenic emissions have extensively been addressed in the scientific literature (Matson et al., 1999, 2002). This is not the case for tropical terrestrial ecosystems where some degree of uncertainty about the influence of increasing Nr deposition remains (Matson et al., 1999, 2002; Galloway et al., 2004; Koehler et al., 2009; Wolf et al., 2011). This increase in tropical ecosystems could have a fertilizing effect and boost ecosystem productivity or decrease it through acidification and related nutrient imbalances (Lewis et al., 2004; Unger et al., 2012). These changes in the biogeochemical cycles are expected to have an impact on ecosystem biodiversity (Matson et al., 1999, 2002; Galloway et al., 2004).

Increased deposition from anthropogenic NO_x emissions in Latin America has been observed not only in the vicinity of the emission sources but also in remote neotropical sites (McDowell et al., 1990; Clark et al., 1998; Phoenix et al., 2006). Nitrogen deposition in the form of NO_3^- has been investigated in tropical lowland sites of Costa Rica (Eklund et al., 1997), Venezuela (Morales et al., 1998), and central Brazilian Amazon (Williams et al., 1997). Eklund et al. (1997) and Williams et al. (1997) found that NO_3^- depositions in Costa Rica and the central Amazon were not contaminated by anthropogenic activities but rather originated from natural sources. Conversely, rain samples from Lake Maracaibo area in Venezuela were strongly contaminated with anthropogenic NO_3^-, potentially originating from industrial and urban upwind emissions (Morales et al., 1998). At high elevation sites, in Costa Rica, Puerto Rico, and Venezuela, cloud water was a significant source of deposition and water input, contributing up to 71% to the total precipitation. NO_3^- concentrations in cloud water were much higher than in rainfall. Samples strongly charged with nitrate ions point to the likely contamination of the airmasses. In most cases the origin of pollutants are thought to be local anthropogenic activities. Long distance transport was not identified as an important deposition source at these locations (Asbury et al., 1994; Gordon et al., 1994a, 1994b; Clark et al., 1998).

Boy et al. (2008) and Fabian et al. (2005, 2009) found, that even in remote areas like the tropical mountain forests of the east Ecuadorian Andes, high nitrogen deposition rates were linked to distant continental anthropogenic upwind-sources from the east. Fabian et al. (2005) stressed the importance of occult precipitation water inputs at the upper parts of the Cordillera and the positive gradient of nitrogen depositions (nitrate and ammonium) with altitude. Both Boy et al. (2008) and Fabian et al. (2005, 2009) observed that deposited nitrate and ammonium were highly correlated, which lead them to the conclusion that they originated from the same source. They pointed out the importance of long-range transport of pollutants for the nutrient budget and identified biomass burning as the most likely source of atmospheric nitrogen deposition to the ecosystem. An explanation is given by Andreae et al. (2004), which studied pyrogenic clouds from biomass burning regions; fires reduce the cloud droplet size, thus delaying precipitation and allowing the polluted air masses to travel greater distances before the pollutants are scavenged by rain. This explains how the polluted air masses may travel as far as the outer rim of the Amazon, where they are finally adiabatically uplifted by the orographic effects when reaching the Andes, causing intense water inputs through impaction of clouds and precipitation (Bendix et al., 2006a, 2006b, 2008a).

In mountain areas, the deposition of long-range transported aerosols can be highly variable depending on location, because of the complex weather patterns generated by the interaction of atmospheric features with topography and the particular exposures of the different landscape features (Lovett and Kinsman, 1990). In tropical mountain forests it is essential to assess the level of pollutant exposure along altitudinal gradients. Depending on location, pollutant loads can vary considerably. Yet, no studies have addressed differences in atmospheric deposition along altitudinal gradients in tropical mountain forests. Likewise, source areas of nitrate precursors related to nitrate deposition have not been investigated in the light of recent developments in remote sensing of tropospheric chemistry and new emission inventories.

We report long term variations of nitrate deposition in precipitation at different altitudes in the *Reserva Biológica San Francisco* (RBSF), in south-east Ecuador. We combined long-term measurements of nitrate concentrations in rain and occult precipitation samples with pollutant transport modeling using emission inventories and satellite data as inputs. Due to the high wind speeds in this area, occult precipitation water here refers to all kind of light precipitation, from wind-driven drizzle down to fog and cloud droplets (Rollenbeck et al., 2007, 2011).

The aim of the current study was to find out (1) how nitrate deposition by occult precipitation and rain water varies in elevation/location along an altitudinal gradient between 1960 and 3180 m a.s.l. and (2) how the relationship between distant upwind emission events and the deposition varies at different elevations/locations.

Additionally, this study contributes to a joint ecological fertilization experiment in southern Ecuador, the Nutrient Manipulation Experiment (NUMEX, Homeier et al., 2012), as part of a joint biodiversity research program (Bendix and Beck, 2009).

2. Study area, data, and methods

To unveil the link between nitrate sources and deposition rates in the RBSF a combination of long-term monitoring of nitrate inputs and nitrogen transport modeling was employed. At the receptor site, in the San Francisco River catchment, *in-situ* data on NO_3^- deposition by rainfall and occult precipitation was collected on a weekly basis, and time series of nitrate concentrations and total deposition rates were compiled.

Regarding the sources of emissions, gridded emission and satellite data are used to model the NO_x transport to yield NO_x daily transport values up to the target coordinates, corresponding to the observation sites in the San Francisco River catchment. This is done by applying trajectory modeling.

The evaluation of data aims at comparing and correlating the observed concentration values and modeled transport values of the observation sites corresponding pixels.

To test the influence of local and synoptic wind exposure on the deposition process, we used local wind observations of the valley and vectorial averages of NCEP/NCAR wind direction at 500 hPa for the corresponding pixels of the observations sites.

The following sections describe the methods for field data sampling and analyses, and the data sets and tools employed for modeling the transport of nitrate precursors to the study area.

2.1. Study site and its climatology

The RBSF (Fig. 1) is located in the south-eastern Andes of Ecuador (4°00′ S and 79°05′ W), on the eastern slope of the *Cordillera Real*, the weather divide between the humid Amazon basin and the dryer interandean valleys west of this *Cordillera* (Beck et al., 2008).