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Atmospheric Chemistry and Physics Discussions

Seasonal cycles of isoprene concentrations in the Amazonian rainforest

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Abstract

Tropical forests are an important global source of volatile organic compounds (VOCs) and other atmospheric trace gases. The high biodiversity in tropical rainforests complicates the extrapolation of biogenic volatile organic compound (BVOC) emissions from leaf-level measurements to landscape and regional or global scales. In Amazônia, a significant fraction of the carbon emitted from the biosphere to the atmosphere is emitted in the form of BVOCs, and the knowledge of these emissions is important to our understanding of tropical and global atmospheric chemistry and carbon cycling. As part of the Large scale Biosphere-atmosphere experiment in Amazônia (LBA). VOC concentrations were measured at two sites near Santarém, Para State, Brazil. The two sites are located in the National Forest of Tapajós, the first corresponding to primary forest and the second to a forest, that was selectively logged. The samples were collected simultaneously at heights of 65 and 55 m (20 and 10 m above forest canopy, respectively). The average isoprene mixing ratio was 2.2–2.5 ppb at the two sites and the standard deviations within a site ranged from 1 to 1.2 ppb. A strong seasonality of isoprene mixing ratio was observed and associated with the wet and dry seasons. The lowest mixing ratios were found during the transition between the wet to dry season, while at the start of the biomass burning season the mixing ratios increase. A qualitative analysis of a one dimensional model demonstrates that the seasonal cycle in concentrations reflects changes in isoprene production by the ecosystem, not changes in boundary layer dynamics or chemistry. The magnitude of the cycle indicates that the physiological capacity of the ecosystem to emit isoprene may depend on water availability although phenological changes could also contribute to the observed variations. A simple 1-D model that assumes mean daytime isoprene fluxes of 1.5 mg $m^{-2}h^{-1}$ and 0.9 mg $m^{-2}h^{-1}$ scaled by an algorithm depending on precipitation at the primary forest and selectively logged sites, respectively, is able to reproduce the observed vertical gradients.

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1. Introduction

Large quantities of volatile organic compounds (VOCs) are emitted into the troposphere from anthropogenic and biogenic sources (Guenther et al., 1995). Global estimates indicate that about 1800 Tg of volatile organic compounds are emitted per year into the atmosphere. Methane is emitted into the atmosphere from both biogenic (natural wetlands) and anthropogenic (domestic ruminants, rice paddies, etc) sources. 510 million Tg yr^{-1} of methane are emitted into atmosphere, with 220 million Tg yr^{-1} emitted by anthropogenic sources. Large quantities of non-methane organic compounds (NMOC), including isoprene (2-methyl-1,3-butadiene), a series of $C_{10}H_{16}$ monoterpenes, $C_{15}H_{24}$ sesquiterpenes, and oxygenated VOCs (methanol, linalool) are also emitted into the atmosphere. Isoprene is emitted at an estimated rate of about 500 Tg yr^{-1} with an additional 130 Tg yr^{-1} of monoterpenes. Guenther et al. (1995) estimate that the global emission of biogenic NMOCs is seven times larger than total anthropogenic emission.

Most biogenic VOCs are emitted from vegetation. Went (1960) was the first to propose that natural emissions of VOCs from trees and other vegetation could have a significant effect on the chemistry of the atmosphere. Often half or more of the biogenic VOCs consist of isoprene (2-methyl-1,3-butadiene) and monoterpenes (e.g. α-pinene, β-pinene, limonene). These compounds are highly reactive in the atmosphere and have short tropospheric lifetimes (~1 h for isoprene). The presence of C=C double bonds in isoprene and monoterpenes makes these compounds highly susceptible to attack by OH, O_3 and NO_3 (Seinfeld and Pandis, 1998). These compounds play an important role in atmospheric chemistry because they contribute to the photochemical production of ozone and the oxidation capacity of the atmosphere. They can also be a source of organic aerosols and contribute to the global carbon cycle. In tropical forest areas, where the concentrations of biogenic VOC are high, the effect of VOC emissions on ozone and OH concentration becomes significant. In the presence of NO_x, hydrocarbons tend to shift the NO to NO_2 balance towards NO_2, thus increasing ozone production, if sufficient NO_x is available (Seinfeld and Pandis, 1998; Atkinson,

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2000). Depending on atmospheric conditions, the degradation of biogenic VOCs can lead to various end-product combinations. The common products of isoprene degradation include formaldehyde, methacrolein and methyl-vinyl-ketone (Seinfeld and Pandis, 1998). Products of α-pinene, which is often the most common monoterpene in the air, include pinonaldehyde, formaldehyde and acetone (Hakola et al., 1994; Nozière et al., 1999).

On a global scale, the largest biogenic VOC emissions are predicted to occur in the tropics with isoprene being the dominant compound emitted, and tropical rainforests are estimated to be the major source of atmopsheric VOCs (Guenther et al., 1995; Zimmerman et al., 1988), due to the combination of high temperatures and large biomass densities. Emissions of isoprene from plants are known to be both light and temperature dependent, and monoterpene emissions are dependent on temperature (Guenther et al., 1991). However, it has been shown that some Mediterranean and tropical plants emit monoterpenes in light and temperature dependent fashion directly from recently fixed carbon (Staudt and Seufert, 1995; Loreto et al., 1996; Kuhn et al., 2001). Seasonal changes in leaf age and leaf biomass density are also expected to influence seasonal variations in BVOC emissions and concentrations (Guenther et al., 1999).

There is a general understanding of the processes driving seasonal variations in isoprene emissions from temperate landscapes (Goldstein et al., 1995; Monson et al., 1994) but very little is known about tropical regions. The few tropical observations that have been reported indicate that seasonal isoprene variations are larger than can be explained by the small seasonal variations in temperature, light and foliage (Guenther et al., 1999; Stefani et al., 2000; Kesselmeier et al., 2002). In this manuscript, we describe the first record of weekly isoprene concentration observations over an annual cycle at a tropical landscape and present an algorithm based on precipitation for predicting seasonal variations in isoprene emissions from tropical landscapes.

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2. Experimental

2.0.1. Site description

The sampling was performed as part of the Large-scale Biosphere atmosphere experiment in Amazonia from December, 2001 through December, 2002 at two sites in the Tapajós National Forest, Pará, Brazil. The first site is located about 7 km west of the Santarém-Cuiabá road (BR-163) at 67 km south of Santarém (2°51.42' S, 54°57.54' W) and is referred to here as site KM67. This sit e is primary rainforest, with closed canopy structure and emergent trees at a height of about 45 m. The samples were collected simultaneously from two heights of the tower at 54 and 64 m using a Teflon tube and a pumping system. The second site is about 4 km from the Santarém-Cuiabá road (BR-163) at 83 km south of Santarém (3° 10,2' S, 54° 58.2' W) and is referred to here as site KM87. This site is a forest where a selective logging project occurred in 2001. The samples were also collected on the tower simultaneously at heights of 54 and 64 m using a Teflon tube. Figure 1 shows the map of Amazonia and the two sampling locations.

2.0.2. VOCs sampling

The samples were collected into stainless steel canisters of 6L volume (BRC/Rasmussen). The canisters were cleaned prior to sampling with a Canister Cleaning System (XonTech, Model 960) during a 4 hour cycle that consisted of 2 moist air cycles (2 h) and 2 dry air cycles (2 h). During the last cycle, the high vacuum pump reduced the pressure in the system to below 10 mtorr. The VOCs were sampled one time per week for each level and in each site. The sample time was between 11:00 am until 02:00 pm, during the period of February to November 2002 at KM83 and December 2001 to November 2002 at KM67. After collection, the samples were sent to the Atmospheric Chemistry Laboratory (LQA) at IPEN ("Instituto de Pesquisas Energéticas e Nucleares") in São Paulo, and then analyzed by GC/MS and GC/FID within 2–3 days

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after sampling.

2.0.3. VOCs analysis

The samples were analyzed with a VARIAN Gas Chromatograph (VARIAN 3600) equipped with a flame ionization detector (FID) and a coupled VARIAN Mass Spectrometer (VARIAN Saturn 2000). The flame ionization detector is used for optimum sensitivity of the VOC species in parallel with a mass selective detection system (GC/MS) for optimum peak identification. The canisters are connected to the analysis system by a sample transfer line heated to 200°C. To concentrate the sample, an aliquot of the air inside the canister is transferred onto a trap immersed in liquid nitrogen with a flow rate of 50 ml min^{-1} for 5 min, giving a total sample volume of 250 ml. The cryogenic preconcentration unit (a stainless steel tube filled with glass beads) is cooled by liquid nitrogen to −180°C, and is thermally desorbed at 200°C for 2 min and injected onto a non-polar DB-1 column (60 m×0,32 mm ID) with a helium carrier gas stream at 2.2 ml min^{-1}. The GC column oven is heated at 6°C min^{-1} from −50°C to 200°C. At the end of the chromatographic column the flow was split, introducing 50% of the flow into the MS systems and 50% into the FID. The FID system is operated at 300°C and is supplied with nitrogen make up gas at 29 ml min^{-1}, hydrogen flow at 30 ml min^{-1} and 300 ml min^{-1} of compressed air flow. The MS system uses an ion trap detector and the system is fully automated and operated by Star Chromatography Workstation Software (version 5.41, VARIAN).

Isoprene quantification was performed by isoprene primary standard (White Martins S.A., Brazil). The range of the calibration curve used was 5 different concentrations between 0.25 ppb and 3.0 ppb. Studies were conducted initially to demonstrate the repeatability of this procedure. The limit of detection (defined as the blank signal plus a signal-to-noise ratio of three) was 0.04 ppb. All the samples were analyzed three times and the reproducibility of the data generally shows a relative standard deviations of about 4% of the concentration range.

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3. Trace gas concentrations

3.1. Primary forest at Tapajós – KM67

The annual precipitation in Tapajós is typically about 2000 mm (Rinne, 2002). The monthly precipitation rates for both sites, Tapajós KM67 and KM83, are shown in Fig. 2. During the period of December 2001 to October 2002 there was a total of 1392 mm of precipitation. The best way to understand the variability of isoprene concentration during this period was to divide it into three seasons: wet, transition and dry season. This division was determined by monthly rain accumulation. The dry season is the period that shows the lowest level of rain, starts in August, and finishes in December. The total dry season precipitation at this station was 42 mm. The wet season is usually between January and May and the transition season occurred during June and July. The transition is characterized by irregular rains, with lower frequency and intensity compared to the wet season. Table 1 shows general meteorological conditions during the dry, transitional and wet seasons at KM67. The mean daytime temperature in Tapajós was almost 28°C; winds were dominant from the E and NE directions with an average of speed about 2.7 m s^{-1}. PAR and temperature were higher in the dry season than in the wet season (Saleska et al., 2004).

The average ozone mixing ratio during the period of maximum radiation (14:00–16:00 LT) in the wet season was 11 ppb. The dry season is when biomass burning occurs. The average mixing ratio correlates with regional biomass burning. The ozone mixing ratio during the dry season without regional biomass burning (August–September) was around 15 ppb, which is close to the wet season value. During December 2001, the average O_3 mixing ratio was 37 ppb during the period of radiation maximum. The 2001 biomass burning season was more intense compared to the 2002 dry season (November–December) when the average O_3 mixing ratio was 26 ppb. A similar pattern was observed for CO mixing ratios. During 2001, the average CO mixing ratios in the wet and dry season without and with biomass burning were 84, 142 and 355 ppb, respectively. In 2002, the average CO mixing ratios in the wet and dry

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season without biomass burning were 105 and 146 ppb, respectively. The biomass burning season occurs during October, November and December, with the more intense period in October and November. But burning begins in August in the western part of the Amazon, and transport will bring some plumes to Tapajós.

Isoprene was the dominant VOC identified by GC/MS and quantified by FID. The isoprene mixing ratio above the canopy at Tapajós KM67 throughout the year is shown in Fig. 3. The mean mixing ratios of isoprene, with standard deviations in parentheses, were 2.2 ppb (1.0) at 54 m and 1.7 ppb (0.9) at 64 m (Table 2). The average decrease in isoprene with height was 41 ppt m^{-1}.

3.2. Forest with selective logging at Tapajós - KM83

The precipitation at this site between February and August 2002 was 960 mm. The precipitation during August was only 2 mm. Table 3 shows the general meteorological conditions in the wet and transition seasons, with August representing the "dry season". The dominant daytime winds were from the E and NE directions during this period, with a wind speed of about 2.8 m s^{-1}. PAR was generally higher in August than during the wet season. The sampling period for this site was between February and December 2002. The isoprene mixing ratios above the canopy at Tapajós KM83 during the campaign are shown in Fig. 3. The average mixing ratio of the isoprene and standard deviations (in parentheses) was 2.5 ppb (1.2) at 54 m and 2.2 ppb (1.2) at 64 m (Table 4). The peak isoprene mixing ratio was about 5.1 ppb and the minimum isoprene mixing ratio was 0.6 ppb. The average decrease in isoprene with height was 20 ppt m^{-1}.

4. Isoprene emissions

Guenther et al. (1995) estimated that midday isoprene fluxes from Amazonian rainforests ranged from about 3 to 7 mg $m^{-2}h^{-1}$. In the Tapajos region, the Guenther et

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al. (1993, 1995) model predicts that emissions should be about 30% higher in the dry season due to higher temperature and light levels. We have developed an additional algorithm to relate isoprene emission to precipitation rates:

$$Ep=2-1.5[P_3/P_{3\,max}], \quad (1)$$

where Ep is a normalized isoprene emission activity factor, P_3 is the average precipitation rate of the past three months and $P_{3\,max}$ is the maximum 3 month precipitation rate. The dimensionless factor Ep is an empirical correction to the basal emission rate used in standard models to predict leaf level isoprene emissions. The application of this factor is analogous to the approach used to account for temporal variations of the basal emission rate due to previous temperature regimes (Geron et al., 2000). The functional form of Ep was chosen to match the observed changes in isoprene concentrations. The algorithm was used to predict weekly variations in isoprene emissions at the Tapajos primary forest site. We combined these isoprene flux estimates with a one dimensional box model to predict boundary layer isoprene concentrations (Jacob and Wofsy, 1988) and surface concentrations using surface to boundary layer average ratios reported by Guenther et al. (1996). The box model balances surface production with storage in the boundary layer and chemical losses. The approach requires knowledge of boundary layer height and isoprene loss rates. Because NO_x levels are low in remote tropical ecosystems, simple HO_x chemistry is assumed. In this study we consider one scenario where the boundary layer height and chemical loss rates are constant and another where boundary layer heights are higher in the dry season (1200–1600 m compared to 500–1000 m, D. Fitzgerald, personal communication) and chemical loss is greater in the dry season due to elevated NO_x and ozone associated with biomass burning. The constant boundary layer height and loss rate scenario is shown as a solid line in Fig. 3. The model results assuming increasing boundary layers and isoprene loss rates in the dry season are shown as a dashed line in Fig. 3. Although there is considerable scatter, the large seasonal variation predicted using Eq. (1) fits the observed pattern reasonably well. The agreement is improved when higher boundary layer and chemical loss rates are assumed for the dry season. It is

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likely that the seasonal variations in isoprene emission rates are driven by changes in the physiological capacity of the ecosystem to emit isoprene and that this is related to water availability. It is also possible that the changes are at least partly due to phenological variations. Additional studies are needed to determine this and to improve the preliminary algorithm given in Eq. (1).

5. Summary and conclusions

Isoprene is an important chemical species in the tropical tropospheric boundary layer but the spatial distribution and seasonal variations are not well known. We observed similar isoprene concentrations at two sites in the Tapajos National Forest in the eastern Amazon basin. At KM67, the mean isoprene mixing ratios were 2.2 ppb and 1.7 ppb, at 54 and 64 m, respectively. At KM83, the mean isoprene mixing ratios were 2.5 ppb and 2.2 ppb, at 54 and 64 m, respectively. The small difference between concentrations at KM67 and KM83 is not significant within the range of uncertainty. However, the concentration at both sites showed significant seasonal variability, with higher isoprene concentrations during dry season. Analysis of a one dimensional budget of isoprene concentrations reveals that this cycle was driven by changes in ecosystem isoprene production. The changes observed cannot be explained by well-established algorithms that account for temperature and light. Guenther et al. (1999) observed a similar pattern of higher isoprene emissions during periods of lower water availability at a tropical forest site in the Congo, Africa and suggested that water status regulates the seasonal variations in the capacity to emit isoprene from tropical ecosystems. This is the most likely cause of the observed seasonal patterns in the Tapajos Forest but we cannot yet rule out phenological changes with season.

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Table 1. Average meteorological conditions and their standard deviations (in parenthesis) calculated during dry, wet and transition season for KM67.

Season *	Temperature (°C)*	PAR (μmol m^{-2} s^{-1})*	Wind speed – 64 m* (m s^{-1})	Wind direction*	Rain (mm)
Wet	26 (1)	817 (342)	2.2 (0.8)	E–NE	1221
Transition	29 (1)	1176 (173)	2.8 (0.7)	E–NE	127
Dry	30 (1)	1159 (213)	2.8 (0.7)	E–NE	42

* Average values performed before and after the sampling interval (11:00–14:00).

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Table 2. Mean isoprene concentration and their standard deviations (in parenthesis) calculated during dry, wet and transition season in Floresta Nacional do Tapajos at 54 m and 64 m for KM67.

Season	Isoprene conc. at 54 m (ppb)	Isoprene Conc. at 64 m (ppb)
Wet	1.9 (1.2)	1.3 (0.8)
Transition	1.4 (0.5)	1.0 (0.4)
Dry	2.8 (0.9)	2.5 (0.8)

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Table 3. Average meteorological conditions and their standard deviations (in parenthesis) calculated during dry, wet and transition season at KM83.

Season *	Temperature (°C)*	PAR (μmol m^{-2} s^{-1})*	Wind speed – 64 m* (m s^{-1})	Wind direction*	Rain (mm)
Wet	30 (3)	768 (262)	2.8 (0.5)	E–NE	721
Transition	32 (1)	1050 (146)	2.9 (0.8)	E–NE	235
Dry	33 (1)	1177 (88)	2.8 (0.8)	E–NE	11

* Average values performed before and after the sampling interval (11:00–14:00).

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Table 4. Mean isoprene concentration (mixing ratio) and their standard deviations (in parenthesis) calculated during dry, wet and transition season in Floresta Nacional do Tapajos at 54 m and 64 m for KM83.

Season	Isoprene conc. at 54 m (ppb)	Isoprene Conc. at 64 m (ppb)
Wet	2.1 (1.3)	1.8 (1.0)
Transition	1.7 (0.3)	1.3 (0.8)
Dry	3.3 (1.0)	2.2 (1.2)

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Fig. 1. Map of the Amazon Basin showing location of the National Forest of the Tapajós at 67 km and 83 km.

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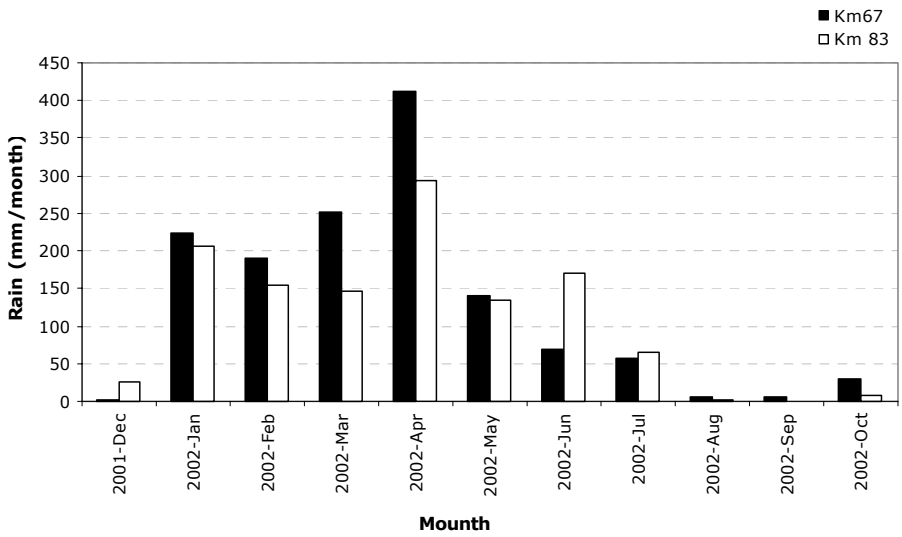

Fig. 2. Cumulative monthly rain at Tapajós KM67 (black) and KM83 (White).

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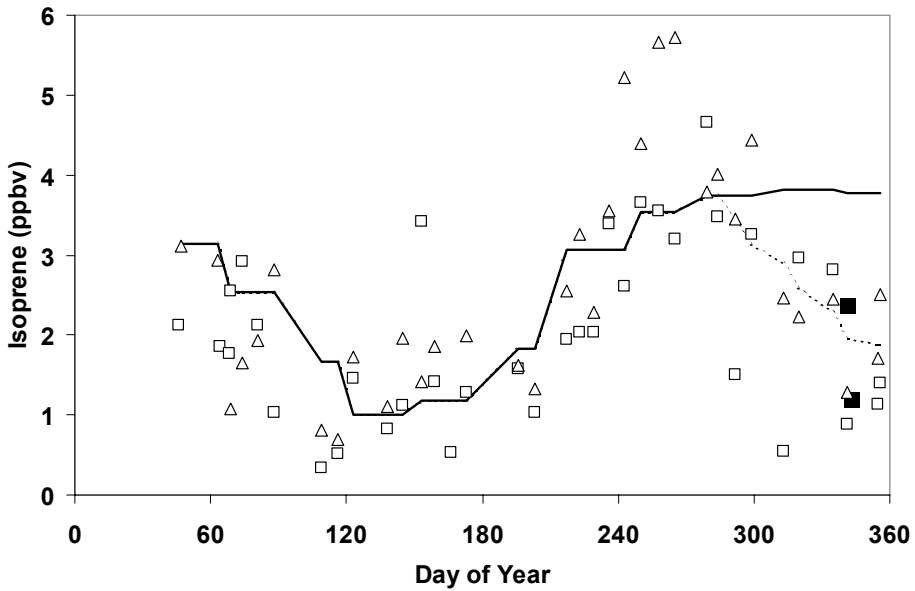

Fig. 3. Average isoprene mixing ratio observed in Floresta Nacional do Tapajos at KM67 (2001: solid squares; 2002: open squares) and KM83 (2002: open triangles) and the fit obtained using the model described in the text assuming a constant mixed boundary layer height (solid line) and a variable mixed boundary layer height (dashed line).

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