

A comparison of dry and wet season aerosol number fluxes over the Amazon rain forest

L. Ahlm¹, E. D. Nilsson¹, R. Krejci¹, E. M. Mårtensson¹, M. Vogt¹, and P. Artaxo²

¹Department of Applied Environmental Science, Stockholm University, Sweden

²Institute of Physics, University of São Paulo, Brazil

Received: 5 November 2009 – Published in Atmos. Chem. Phys. Discuss.: 15 December 2009

Revised: 12 March 2010 – Accepted: 19 March 2010 – Published: 31 March 2010

Abstract. Vertical number fluxes of aerosol particles and vertical fluxes of CO₂ were measured with the eddy covariance method at the top of a 53 m high tower in the Amazon rain forest as part of the LBA (The Large Scale Biosphere Atmosphere Experiment in Amazonia) experiment. The observed aerosol number fluxes included particles with sizes down to 10 nm in diameter. The measurements were carried out during the wet and dry season in 2008. In this study focus is on the dry season aerosol fluxes, with significant influence from biomass burning, and these are compared with aerosol fluxes measured during the wet season.

Net particle deposition fluxes dominated in daytime in both seasons and the deposition flux was considerably larger in the dry season due to the much higher dry season particle concentration. The particle transfer velocity increased linearly with increasing friction velocity in both seasons. The difference in transfer velocity between the two seasons was small, indicating that the seasonal change in aerosol number size distribution is not enough for causing any significant change in deposition velocity. In general, particle transfer velocities in this study are low compared to studies over boreal forests. The reasons are probably the high percentage of accumulation mode particles and the low percentage of nucleation mode particles in the Amazon boundary layer, both in the dry and wet season, and low wind speeds in the tropics compared to the midlatitudes.

In the dry season, nocturnal particle fluxes behaved very similar to the nocturnal CO₂ fluxes. Throughout the night, the measured particle flux at the top of the tower was close to zero, but early in the morning there was an upward parti-

cle flux peak that is not likely a result of entrainment or local pollution. It is possible that these morning upward particle fluxes are associated with emission of primary biogenic particles from the rain forest. Emitted particles may be stored within the canopy during stable conditions at nighttime, similarly to CO₂, and being released from the canopy when conditions become more turbulent in the morning.

1 Introduction

The Amazonian forest is the largest tropical forest on Earth. During the wet season, the atmospheric boundary layer over the Amazon is relatively clean with low aerosol number concentrations (Artaxo et al., 2002; Martin et al., 2010; Zhou et al., 2002). In the dry season, however, when biomass burning is no longer suppressed by intense precipitation, aerosol concentrations are considerably higher and the aerosol population is dominated by anthropogenic particles (Andreae et al., 1988; Artaxo et al., 1998; Bowman et al., 2009). Elevated particle concentrations in the dry season influence climate directly through increased scattering of incoming solar radiation which in turn may affect the photosynthetic rate and thereby the regional carbon balance (Oliveira et al., 2007). Additionally, biomass burning particles are efficient cloud condensation nuclei (CCN) and therefore influence the formation of clouds and precipitation (Andreae et al., 2004; Gunthe et al., 2009; Koren et al., 2008). Moreover, absorption of solar radiation by smoke particles may lower the relative humidity and increase temperature in the absorbing layer, thereby reducing cloudiness and changing the atmospheric stability profile (Ackerman et al., 2000), which in turn affects turbulent fluxes of heat, moisture and even



Correspondence to: L. Ahlm
(lars.ahlm@itm.su.se)

aerosol particles. Because of the intense convective activity over the rain forest, often associated with the Intertropical Convergence Zone (ITCZ), natural and anthropogenic aerosols can be uplifted to higher altitudes and be transported far away from the tropics and in this manner also have a global impact on climate (Andreae et al., 2001).

In order to fully represent the impact from biomass burning on regional and global climate, it is important to reduce the uncertainties in particle number emission factors from biomass burning (Andreae and Merlet, 2001; Lohmann et al., 2007), but also to understand the processes controlling removal of aerosols from the atmosphere. The most important deposition processes are wet and dry deposition. The efficiency of dry deposition is highly dependent on particle size (Slinn et al., 1982). Particle emission from biomass burning is dominated by accumulation mode particles (Artaxo et al., 1994; Reid et al., 2005), for which there is no efficient dry deposition mechanism.

Rissler et al. (2004) investigated the surface aerosol size distribution in Balbina, located 125 km northeast of Manaus, and found that the size distribution was dominated by an Aitken and an accumulation mode both in the dry and wet season. In the same study, particle concentrations were elevated during an aged biomass burning period compared to the clean background air mass by nearly a factor of 2 in the Aitken mode size range, and 4–5 times in the accumulation mode size range. Thus, a higher percentage of accumulation mode particles may be expected in the dry season compared to the wet season, since biomass burning is active primarily in the dry season. Furthermore, reduced wet removal of accumulation mode particles during transition from wet to dry season will also result in an increasing percentage of accumulation mode particles. This percentage increase could have an impact on the average dry deposition velocity. By measuring vertical aerosol number fluxes, the dry deposition sink can be quantified. Furthermore, vertical particle fluxes reveal whether the rain forest always acts as a net particle sink, or if it under certain conditions may be a net particle source. Natural biogenic particles are present in the Amazon basin in both the dry and wet season. The coarse aerosol fraction is dominated by primary biogenic aerosol particles (Graham et al., 2003). However, the contribution of primary aerosol emission to the fine aerosol fraction is more uncertain. Several biogenic related elements (e.g. K, P, S, Zn) in plants are present in the fluids circulating in the plants and can be released from the plant during transpiration (Nemeruyk, 1970). Fish (1972) suggested that haze observed over forests could be due to submicrometer particles from electrical generation of biogenic aerosol by leaves. Furthermore, decaying vegetation may produce aerosol particles (Schnell and Vali, 1973). Fungal spores are usually in the diameters size range 1–30 μm (Jones and Harrison, 2004), suggesting that they do not contribute significantly to the aerosol number population. However, the number of fungal spores existing on Earth is assumed to be in the range

of 1–1.5 million (Elbert et al., 2007), but only about 40 000 are well-characterized (Rossman, 1994), why it cannot be excluded that fungal spores also makes a significant contribution to the fine mode. Finally, bacteria are typically 0.25–8 μm in diameter (Thompson, 1981) and may therefore make a contribution also in the sub-micron range.

To our knowledge, Ahlm et al. (2009) contains the first peer-review published results ever on eddy covariance aerosol particle fluxes over the Amazon rain forest. That study was based on wet season measurements in the Cuieiras Ecological Reserve close to Manaus in the Northern part of the Amazon rain forest. The study showed that net particle fluxes pointed downward even in the absolute cleanest conditions. This was an indication that the contribution from primary aerosol emission may be low in the wet season.

In this study, focus is on the dry season aerosol number fluxes, with larger impact from anthropogenic sources, and these fluxes are compared with particle fluxes measured in the wet season. The goal is to quantify the dry deposition sink and also to investigate whether the particle deposition velocities change during transition from the wet season into the dry season. Furthermore, it is tested whether the rain forest is a net sink of particles also in the dry season, or if particle emission from the surface under certain circumstances may dominate over the dry deposition sink. This Brazilian-Swedish project AMAFLUX (Amazonian Biosphere-Atmosphere Aerosol Fluxes in view of their potential control of cloud properties and climate) was carried out as a part of the larger international project LBA (The Large Scale Biosphere Atmosphere Experiment in Amazonia) and the measurement were performed in 2008.

2 Method

2.1 Site description

The measurements were carried out at the top of the 53 m high tower K34 in the Reserva Biológica do Cuieiras (2°35.37' S, 60°06.92' W), approximately 60 km NNW of Manaus, Brazil. The tower is a research facility operated by INPA (The Brazilian National Institute for Research in Amazonia). The canopy height in the Cuieiras Reserve is between 30 and 35 m (Kruijt et al., 2000). Figure 1 shows the location of the measurement site. A more detailed description of this site can be found in Ahlm et al. (2009).

2.2 Eddy covariance measurements

The eddy covariance method was used to measure the mean vertical turbulent aerosol number flux $\overline{N'w'}$, where N' and w' represents fluctuations in aerosol number concentration and vertical wind speed from the temporal means of these parameters, and the cross bar represents a temporal mean of the product of the two fluctuations.

Campos et al. (2009) investigated turbulent time scales at K34 by using a multiresolution decomposition technique. They found that the average time scale was below 200 s at nighttime and below 1200 s in daytime for CO₂ and energy fluxes. Hence, it is preferable to use short time scales when rotating and de-trending fluxes measured within the nocturnal boundary layer to obtain as stationary conditions as possible and thereby minimizing the uncertainty of the flux. However, in daytime it is necessary to use longer time scales to include the largest eddies within the mixed layer. Even though the daytime turbulence time scale is on average below 1200 s, eddies with considerably lower frequencies have been observed to contribute to energy fluxes over the Amazon (Finnigan et al., 2003). However, the variability of the aerosol number concentration is much larger than the variability of temperature and water vapor (or even CO₂). To de-trend particle concentrations and calculate the particle fluxes over very long time scales would often produce large errors and increase the uncertainty of the particle flux.

For this study the vertical aerosol flux $\overline{N'w'}$ was calculated and linearly de-trended over three different time scales to make it possible to investigate both daytime and nighttime fluxes. The chosen time scales were 30, 10 and 3 min long. The aerosol data was shifted in relation to the wind data to correct for the time lag in the sampling line (calculated from the maximum correlation). Turbulent fluxes of momentum, energy and CO₂ were calculated in a similar way, but only over time scales of 30 min since the magnitude of these fluxes is not the main objective of this study.

2.3 Instrumentation

2.3.1 Flux measurements

The 3-D wind components and temperature were measured with a Gill Windmaster ultrasonic anemometer, and logged at 20 Hz. To measure the total aerosol number concentration (particle diameter $D_p > 10$ nm) we used a Condensation Particle Counter (CPC), model TSI 3010, which was logged at 1 Hz. The aerosol was sampled just beneath the sonic head through a 4 m long 1/4-inch stainless steel sampling line. The sampling flow through the CPC was 1.08 l min⁻¹.

Concentrations of CO₂ and H₂O were measured by a Li-7500 Open Path Analyzer. The Licor was logged both as digital RS232 signals through an EDG-4508 gateway and as analog signals through the Gill windmaster auxiliary input channels, in both cases at 20 Hz.

2.3.2 Aerosol number size distribution measurements

Aerosol number size distributions were measured with a SMPS (Scanning Mobility Particle Sizer) system. The SMPS system included a DMA (Differential Mobility Analyzer) of model TSI 3081, an electrostatic classifier of model TSI 3080, and a CPC of model TSI 3010. Aerosol number con-

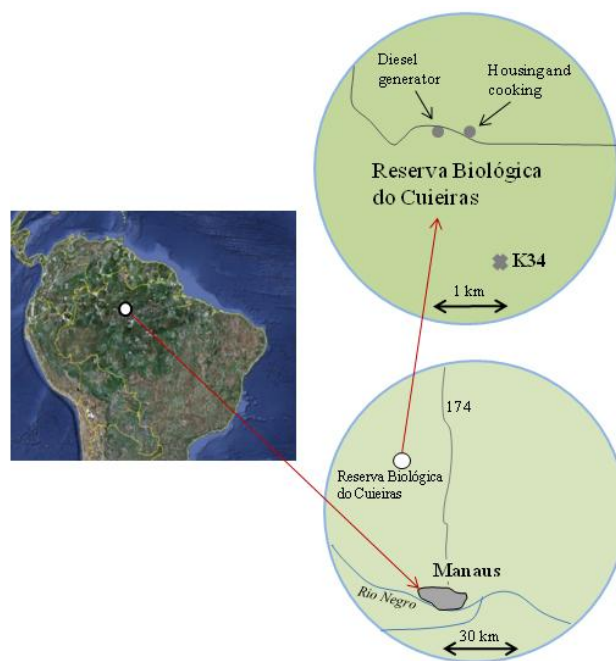


Fig. 1. Overview map of the measurement site in the Reserva Biológica do Cuieiras. The map over northern South America to the left is taken from Google Earth.

centrations were measured in 95 size bins in the particle diameter interval 10 to 300 nm. The sampling time was 5 min and the flow rate was 1 lpm.

2.3.3 Additional data used during data analysis

Mass concentration of equivalent black carbon (BC_e) was provided by São Paulo University using a Multi-Angle Absorption Photometer (MAAP). This measurement derives the concentration of BC_e (Andreae and Gelencsér, 2006) from the determination of light absorption at a wavelength of 670 nm using an empirical mass absorption efficiency of 6.5 m² g⁻¹. BC_e was measured at a container close to the house at the center of the research station, approximately 2 km north of K34.

Additional meteorological parameters (temperature, relative humidity, rain amount and photosynthetic active radiation) were measured at the K34 tower and provided by INPA. These were logged on a Campbell CR-10 (Campbell Scientific UK) data logger with a sampling interval of 30 s and stored as either 10 or 30 min averages.

2.4 Flux corrections and random uncertainty

The eddy covariance method requires stationary conditions. In this study, the nonstationarity test by Foken and Wichura (1996) was applied to the particle fluxes measured over 30 min, in order to filter out fluxes measured in non-stationary conditions. The averaging period 30 min was

divided into sub-periods of 5 min. If the difference between the flux calculated over 30 min and the mean of the covariances calculated over the 5 min intervals was larger than 60% (Järvi et al., 2009), the flux was rejected. A fraction of 64% of the particle fluxes passed the instationarity test.

Particle fluxes measured with the eddy covariance method are underestimated due to the limited time response of the CPC and attenuation of turbulent fluctuations in the sampling line. The frequency first order response time constant τ_c of the TSI 3010 has been estimated to 0.8 s (Doebelin, 1990). A total τ_c for both CPC and sampling line was estimated to 1.3 s by using transfer equations for damping of particle fluctuations in laminar flow (Lenschow and Raupach, 1991) and in a sensor (Horst et al., 1997). The aerosol fluxes in this study have been corrected for these fluctuation attenuations according to Horst et al. (1997). The correction was on average 16% of the measured net aerosol flux in the dry season and 15% in the wet season.

In this study, the Webb correction has been applied to the CO₂ and the latent heat flux. The Webb correction at noon (when energy fluxes are at maximum) reduced the net downward CO₂ flux with about 45% in the dry season and 25% in the wet season. The corresponding increase in latent heat flux was 13% and 9% in the dry and wet season, respectively. The Webb correction has not been applied to the particle fluxes. The motivation for this can be found in Ahlm et al. (2009).

The random uncertainty in flux δF can be expressed as (Wyngaard, 1973):

$$\delta F = \sqrt{\frac{2\tau}{T} \left[(w'N')^2 - w'N'^2 \right]} \quad (1)$$

where T is the averaging period, and τ is the integral time scale, in this study estimated according to Rannik et al. (2009).

3 Results and discussion

The flux measurements included in this study were performed between 12 March and 18 May (wet season) and between 15 July and 12 August 2008 (dry season). Concerning the wet season CPC measurements, 37% of the data had to be removed because of technical problems, mainly linked to water uptake in the CPC butanol reservoir. The corresponding loss of data from the dry season was only 8%.

Of the CO₂ and H₂O measurements, 15% of the data were rejected from the wet season data and 19% from the dry season data, primarily due to problems with electricity or computer software and spikes in raw data during rainfall.

Meteorological and BC_e measurements ran more or less continuously during the two flux measurement periods. The intention was to measure aerosol number size distributions in parallel with the aerosol flux measurements. However, due to technical failure aerosol number size distributions have only been measured in a separate period between 13 June and 7

July, just before the period of the dry season flux measurements.

3.1 Average conditions during the campaign

Tables 1–2 show the average meteorological conditions, concentrations and fluxes during the two measurement periods, the dry and wet season, respectively. The flux parameters are defined as positive when the flux is upward and negative when the flux is downward.

The difference in BC_e concentration between the dry and wet season (Tables 1–2) in this study shows the impact of biomass burning emissions in the dry season at the Cuieiras Reserve. The mean dry season BC_e concentration was $259 \pm 115 \text{ ng m}^{-3}$ and the corresponding concentration in the wet season was $80 \pm 45 \text{ ng m}^{-3}$ (mean \pm standard deviation). The other parameters will be discussed closer in next section.

3.2 Diurnal cycles of meteorological parameters

This section deals with the average diurnal cycles of meteorological parameters. These are important when later interpreting the vertical aerosol number fluxes. The diurnal cycles (Fig. 2a–j) are shown as medians of half-hour mean values. The reason for choosing median cycles instead of mean cycles is to reduce the weight of extreme values and instead show what is typically happening. The only exception is the diurnal cycle of rainfall (Fig. 2j), where it makes more sense to use mean cycle, since the median rain amount is zero for a large fraction of the half hour intervals forming the diurnal cycle.

The sunrise was around 06:00 LT (local time) and the sunset at 18:00 LT, which can be seen in the curve showing Photosynthetic Active Radiation (PAR) (Fig. 2a). The PAR is higher in the dry season than in the wet season, due to less cloudiness in the dry season. The curves for sensible (Fig. 2b) and latent (Fig. 2c) heat fluxes are rather well correlated with the PAR, and these fluxes are larger in the dry season because the incoming solar radiation (as well as the PAR) then is higher. However, the sensible and latent heat fluxes start to increase first ~ 1.5 h after sunrise. This delay might be an effect of negative radiation balance also a while after sunrise.

Also the temperature (Fig. 2d) is higher in the dry season with the largest difference between the two seasons prevailing during the afternoon.

As discussed by Ahlm et al. (2009), the characteristics of the tropical boundary layer and the mechanisms governing its evolution can be revealed by investigating the diurnal cycle of water vapor concentration (Fig. 2e). In the morning, before the nocturnal inversion has been dissipated, the mixed layer grows very slowly and the water vapor from evapotranspiration is trapped in a thin mixed layer connected to the surface. However, after the nocturnal inversion has been dissipated and resistance to further growth is much lower (Stull

Table 1. Average conditions for critical parameters of measurements in the dry season. The \pm range after the mean value is the standard deviation and the numbers after the median are 10 and 90 percentiles. The average diurnal maximum and minimum have been calculated by taking the median value of all diurnal maxima and minima throughout the campaign. The numbers within the brackets in the max and min columns are 10 and 90 percentiles.

	Mean	Median	Diurnal max	Diurnal min
Temperature ($^{\circ}\text{C}$)	26.4 \pm 3.0	26.0 (22.3, 28.9)	30.8 (28.9, 32.2)	22.9 (21.9, 23.9)
Relative humidity (%)	74.6 \pm 14.6	76.2 (52.8, 92.8)	93.4 (82.9, 96.1)	52.0 (44.8, 67.5)
Rain amount per day (mm)	2.8 \pm 5.9	0.2 (0, 10.1)	–	–
Photosynthetic active radiation (Wm^{-2})	130.6 \pm 179.7	3.4 (0, 433.9)	540 (434, 603)	0
Sensible heat flux (Wm^{-2})	19.9 \pm 44.4	–0.2 (–9.2, 91.5)	146.9 (80.3, 196.8)	–24.6 (–60.8, –10.0)
Water vapor molar density (mmol m^{-3})	1242 \pm 125	1268 (1098, 1363)	1359 (1306, 1433)	1046 (628, 1190)
Latent heat flux (Wm^{-2})	90.7 \pm 144.1	11.9 (–2.2, 312.9)	429 (209, 527)	–9.6 (–106.4, –2.0)
Wind speed (ms^{-1})	2.2 \pm 0.9	2.1 (1.1, 3.3)	4.0 (3.2, 5.3)	0.7 (0.3, 1.4)
Friction Velocity (ms^{-1})	0.19 \pm 0.17	0.14 (0.03, 0.46)	0.59 (0.37, 0.72)	0.011 (0.006, 0.026)
Inverted Obukhov length 1/L (m^{-1})	3.5 \pm 112.2	0.01 (–0.07, 0.24)	1.87 (0.47, 29.15)	–0.76 (–22.31, –0.06)
CO ₂ molar density (ppm)	368 \pm 18	362 (353, 388)	401 (383, 467)	352 (345, 356)
CO ₂ flux ($\mu\text{mol m}^{-2} \text{s}^{-1}$)	–1.46 \pm 6.32	0.09 (–11.05, 5.14)	11.1 (7.1, 19.5)	–15.0 (–19.0, –7.3)
Particle number concentration (cm^{-3})	1513 \pm 721	1352 (869, 2292)	2388 (1247, 4172)	982 (513, 1363)
Particle number flux ($10^6 \text{m}^{-2} \text{s}^{-1}$)	–0.45 \pm 3.89	–0.20 (–2.66, 1.72)	4.11 (0.91, 16.6)	–5.14 (–24.79, –1.71)
BC _e concentration (ng m^{-3})	259 \pm 115	245 (141, 375)	453 (250, 868)	146 (53, 202)

Table 2. Average conditions for critical parameters of measurements in the wet season. The \pm range after the mean value is the standard deviation and the numbers after the median are 10 and 90 percentiles. The average diurnal maximum and minimum have been calculated by taking the median value of all diurnal maxima and minima throughout the campaign. The numbers within the brackets in the max and min columns are 10 and 90 percentiles.

	Mean	Median	Diurnal max	Diurnal min
Temperature ($^{\circ}\text{C}$)	24.6 \pm 2.3	24.0 (22.2, 28.2)	28.9 (26.3, 31.0)	22.2 (21.6, 23.1)
Relative humidity (%)	86.4 \pm 10.6	90.7 (69.2, 95.9)	96.0 (94.7, 96.5)	66.1 (54.6, 78.6)
Rain amount per day (mm)	10.8 \pm 12.8	5.6 (0.2, 29.1)	–	–
Photosynthetic active radiation (Wm^{-2})	84.6 \pm 129.7	1.5 (0, 303.6)	455 (275, 537)	0
Sensible heat flux (Wm^{-2})	14.4 \pm 47.2	–0.7 (–11.7, 77.3)	142.7 (36.0, 230.6)	–26.7 (–102.6, –10.2)
Water vapor molar density (mmol m^{-3})	1120 \pm 170	1140 (970, 1250)	1246 (1160, 1606)	834 (275, 1150)
Latent heat flux (Wm^{-2})	71.4 \pm 133.1	11.5 (–2.2, 254.8)	368 (166, 489)	–23.1 (–116.2, –0.35)
Wind speed (ms^{-1})	2.0 \pm 0.9	1.9 (0.9, 3.0)	3.6 (2.7, 4.8)	0.4 (0.2, 1.2)
Friction Velocity (ms^{-1})	0.21 \pm 0.16	0.18 (0.04, 0.42)	0.52 (0.35, 0.77)	0.018 (0.008, 0.063)
Inverted Obukhov length 1/L (m^{-1})	0.03 \pm 0.94	0.01 (–0.05, 0.11)	0.76 (0.05, 6.05)	–0.31 (–6.2, –0.01)
CO ₂ molar density (ppm)	392 \pm 41	384 (366, 422)	430 (398, 540)	364 (349, 372)
CO ₂ flux ($\mu\text{mol m}^{-2} \text{s}^{-1}$)	–1.38 \pm 7.37	0.47 (–13.15, 6.06)	11.3 (4.2, 17.3)	–17.6 (–19.7, –11.2)
Particle number concentration (cm^{-3})	682 \pm 780	466 (243, 1260)	853 (445, 5338)	263 (133, 458)
Particle number flux ($10^6 \text{m}^{-2} \text{s}^{-1}$)	–0.32 \pm 3.50	–0.10 (–1.44, 1.03)	1.70 (0.40, 14.3)	–2.41 (–20.2, –0.51)
BC _e concentration (ng m^{-3})	80 \pm 45	69 (36, 140)	131 (77, 263)	33.5 (21.4, 64.1)

et al., 1988), the mixed layer grows fast and entrainment of drier air from above then dominates over evapotranspiration. This results in decreasing water vapor concentration despite enhancing evapotranspiration with increasing PAR. In Fig. 2e, it is obvious that the switch from increasing to decreasing water vapor concentration occurs around 09:00 LT both in the dry and in the wet season. Hence, it seems that

the burning off of the nocturnal inversion occurs at approximately the same time in the two seasons. The water vapor concentration is generally higher in the dry season than in the wet season even though the relative humidity (Fig. 2f) is higher in the wet season (due to lower temperature in the wet season).

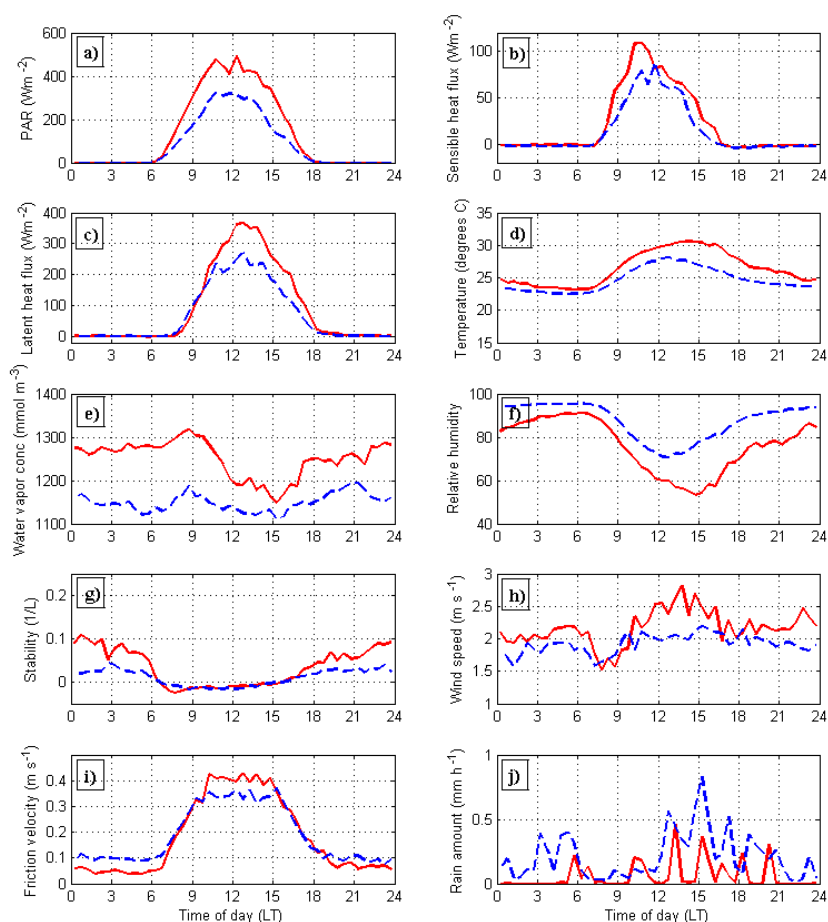


Fig. 2. Meteorological parameters measured at the top of the K34 tower. Solid red lines represent dry season and dashed blue lines represent wet season for (a) photosynthetic active radiation (PAR), (b) sensible heat flux, (c) latent heat flux, (d) temperature, (e) water vapor concentration (f) relative humidity, (g) stability (L^{-1}), (h) horizontal wind speed, (i) friction velocity, and (j) rain amounts.

Figure 2g reveals the differences in stability, L^{-1} , between the two seasons, where L is the Obukhov length. In daytime, an unstable convective boundary layer is present both in the dry and wet season with similar values on L^{-1} . However, the difference in stability between the two seasons is clearly visible at nighttime. A typical nocturnal stable boundary layer forms one or two hours before sunset in the dry season (also seen as negative sensible heat flux in Fig. 2b) and at nighttime the stratification is highly stable. The nocturnal boundary layer reaches on average a final depth of 80–180 m (Garstang et al., 1990). However, this stable nocturnal layer is less pronounced in the wet season with sometimes unstable conditions also at nighttime, seen in higher nighttime wet season friction velocities (Fig. 2i) and also higher nighttime wet season rain amounts (Fig. 2j). The daytime friction velocity is often higher in the dry season than in the wet season, probably to a large extent due to higher daytime wind speeds (Fig. 2h). The friction velocity starts to increase rapidly first one hour after sunrise. The top of the canopy needs to be warmed up before the air temperature above the canopy starts

to increase. When the unstable layer reaches the top of K34, about 20 m above the canopy, the friction velocity at the top of K34 is expected to increase. However, since the nocturnal layer is 80–180 m it seems to last until 09:00 LT until the whole nocturnal layer is dissipated (Fig. 2e).

3.3 Diurnal cycles of concentration and flux of CO_2

In this section, diurnal cycles of CO_2 concentration and flux are analyzed. The primary reason for investigating also fluxes of CO_2 in this study, is that the diurnal cycles of the CO_2 and the particle flux show some similarities that will be a help when interpreting the particle fluxes in detail in Sect. 3.4.

The CO_2 concentration and flux have a very distinct diurnal cycle (Fig. 3). In daytime there is an uptake of CO_2 by the forest (downward fluxes) and the atmospheric CO_2 concentration consequently decreases. During evening and nighttime, when there is no photosynthetic active radiation and only CO_2 emission (upward fluxes), instead the CO_2

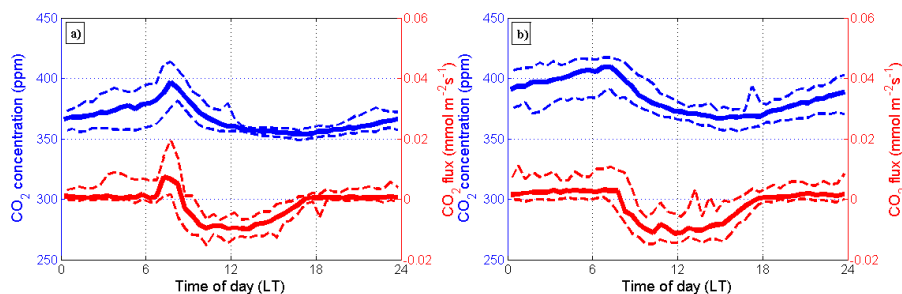


Fig. 3. Median diurnal cycles of CO₂ vertical flux (solid red lines) and concentration (solid blue lines) in the dry season (a) and wet season (b). Dashed lines are 25 and 75 percentiles.

concentration increases. An interesting difference between the two seasons is the peak in upward flux, between 07:00 LT and 08:00 LT, apparent in the dry season curve but not in the wet season curve. In the same time interval, the dry season concentration rapidly increases followed by a peak in concentration, whereas the wet season concentration curve has a more continuous shape. Malhi et al. (1998) noticed that on calm nights with stable stratification, most of the respired CO₂ is stored within the forest canopy and released in the morning when conditions become more turbulent, while during less stable nights most of the CO₂ is released intermittently throughout the night. They found the threshold friction velocity, separating the two cases, to be 0.1 ms⁻¹. This explains the patterns seen in Fig. 3. In the wet season, nighttime friction velocities (Fig. 2i) are close to 0.1 ms⁻¹ and the CO₂ flux points steady upward throughout the night, although varying in magnitude. In the dry season, however, nighttime friction velocities are considerably lower and the nighttime CO₂ flux is therefore close to zero with a following large emission peak at 07:00–08:00 LT when conditions become more turbulent.

It has long been known that respiration is often underestimated by nighttime eddy covariance measurements over forest canopies and that this underestimation is most significant in calm nights with low wind speeds (Goulden et al., 1996), a very frequent situation. At nighttime, the canopy layer becomes decoupled from the atmosphere above. The airflow above the canopy is then synoptically driven, while the airflow within the canopy is dominated by orographic effects, in this case leading to mainly local katabatic flows (Aubinet et al., 2003; Marcolla et al., 2005). There is growing evidence that nighttime advection caused by these drainage flows is the root cause of the failure to capture the respiration flux in stable conditions at nighttime (Finnigan et al., 2008).

Araújo et al. (2008) investigated the nocturnal CO₂ concentration field in the heterogeneous terrain of the Cuieiras Reserve of valleys and slopes and found that, particularly during stable nights, large amounts of CO₂ were transported downslope by drainage flows from the K34 plateau and being accumulated in valleys. This is useful information when

later discussing the diurnal cycle of the vertical particle flux in Sect. 3.4.4.

3.4 Aerosol number fluxes and concentrations

3.4.1 Aerosol number size distribution

The aerosol number size distributions were measured just before the dry season flux measurement period and are therefore representative of the dry season size distribution. Figure 4a shows the median aerosol number size distributions during the period. The vertical bars represent 25 and 75 percentiles. Numbers of nucleation mode particles are low and the size distribution is dominated by an accumulation mode, centered at a diameter of ~150 nm. The Aitken mode is most evident in the 25 percentile curve, but can be observed also in the median curve, and is centered at a diameter of ~70 nm. The so called Hoppel-minimum (Hoppel et al., 1994), separating the two modes, is located at a diameter of ~100 nm. Particles larger than ~100 nm are easily activated in clouds over the Amazon basin and can thereby be cloud-processed and grow efficiently (Rissler et al., 2004), which explains the minimum between the two modes. The reason that the Hoppel minimum is most apparent in the 25 percentile curve in Fig. 4 may be explained by that lower particle concentrations are associated with days with more wet deposition and thereby more clouds with potentially more cloud-processing of aerosol particles.

Zhou et al. (2002) investigated the wet season aerosol number size distribution at Balbina, located 125 km northeast of Manaus, relatively close to the site of this study. They described the wet season size distribution by an accumulation mode, an Aitken mode and a nucleation mode with geometrical mean diameters of 151, 68 and 24 nm, respectively. The geometrical mean diameters of the Aitken and the accumulation modes in that study are very close to the diameters of the observed modes in Fig. 4a.

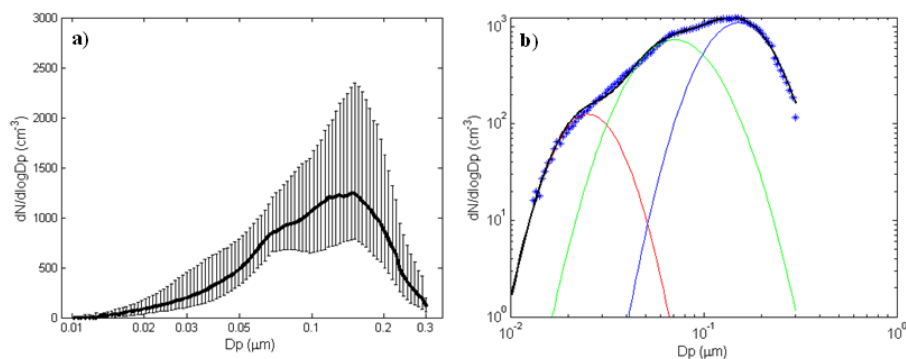


Fig. 4. (a) Median aerosol number size distribution (solid line) with vertical bars representing 25 and 75 percentiles during the period 13 June to 7 July measured with the SMPS system. (b) The median aerosol size distribution (blue stars) described with three modes (red, green and blue lines) resulting from a log-normal fitting. The black solid line represents the sum of the three modes.

In Fig. 4b, it is shown that the median size distribution of this study can be described by three modes, an accumulation mode, an Aitken mode and a nucleation mode, similarly to the size distribution in Zhou et al. (2002). The number concentrations, geometrical mean diameters, and geometrical standard deviation of the three modes in both this study, representing the dry season, and in the study by Zhou et al. (2002), representing the wet season, are provided in Table 3. Apart from the obvious difference between the two seasons that the number concentrations are much higher in the dry season, Table 3 also reveals that the percentage of accumulation mode particles are higher in the dry season than in the wet season. As was discussed in the introduction, this is logical since biomass burning is known to be a large source of accumulation mode particles (Reid et al., 2005), and biomass burning is active primarily in the dry season. Furthermore, wet deposition is an efficient sink of accumulation mode particles, and decreasing precipitation in the dry season therefore increases the lifetime of accumulation mode particles.

The percentage of nucleation mode particles is lower in the dry season than in the wet season, despite the fact that the nucleation mode has been defined as wider in the dry season, according to the geometrical mean diameters in Table 3. The lower percentage of nucleation mode particles and higher percentage of accumulation mode particles in the dry season should have a damping effect on the average particle deposition velocity in the dry season compared to the wet season.

3.4.2 Concentrations of particles in the dry and wet season

The mean aerosol number concentration and standard deviation in the dry and wet season periods were $1513 \pm 721 \text{ cm}^{-3}$ and $682 \pm 780 \text{ cm}^{-3}$, respectively (Tables 1–2). The corresponding median values were 1352 cm^{-3} and 466 cm^{-3} . Hence, the mean particle concentration was roughly two

Table 3. Statistics of the aerosol number size distributions in the wet season in March and April at Balbina (Zhou et al., 2002) and in the dry season in June and July in the Cuieiras Reserve (this study).

Mode	Number Concentration (cm^{-3})	Geometrical Mean Diameter (nm)	Geometrical Standard Deviation (nm)
Wet Season (Zhou et al., 2002)			
Accumulation	146	151	1.40
Aitken	200	68	1.40
Nucleation	48	24	1.31
Dry Season (this study)			
Accumulation	421	151	1.41
Aitken	322	71	1.49
Nucleation	42	25	1.36

times higher in the dry season than in the wet season while the median particle concentration was approximately three times higher in the dry season. This means that the dry season particle concentration was typically three times higher than the wet season concentration, but some occasionally high peaks in wet season particle concentration brings the mean concentrations in the two seasons closer to each other compared to the median concentrations. This can also be seen in the higher standard deviation in the wet season aerosol number concentration.

The difference in particle concentration between the two seasons is much less pronounced in this study than in other studies in Rondônia in the southwestern part of the Amazon rain forest (Rissler et al., 2006). The reason for this is that the Cuieiras Reserve is located in an area of pristine rain forest where the direct influence of biomass burning is much lower than in Rondônia or other locations in the southern part of the Amazon rain forest. Even in the dry season, impact of biomass burning emissions is not very high at the Cuieiras Reserve, but can be observed most of the time.

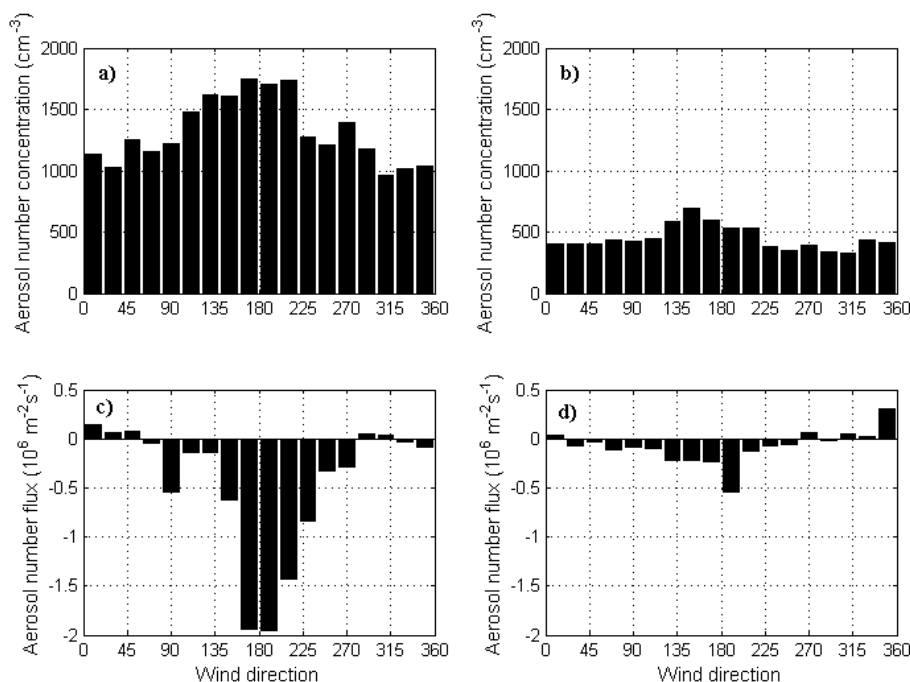


Fig. 5. Dependence on wind direction for the aerosol number concentration in the dry (a) and wet (b) season and for the aerosol number flux in the dry (c) and wet season (d).

3.4.3 Dependence on wind direction

Figure 5 shows the dependence on wind direction for the aerosol number concentration in the dry (Fig. 5a) and wet (Fig. 5b) season and for the aerosol number flux in the dry (Fig. 5c) and wet (Fig. 5d) season. The dry season aerosol number concentration peaks when the wind direction is between 170–200 degrees, which represents advection of air with large influence from biomass burning in the southern part of the Amazon rain forest. The wet season aerosol concentration peaks when winds are southeasterly which represents advection from the city Manaus. Hence, it seems that Manaus is the dominant source of air pollution in the wet season but not in the dry season.

In Fig. 5c–d it is obvious that downward particle fluxes dominate both in the dry and wet season and deposition fluxes are considerably larger in the dry season when particle concentrations are much higher. The net upward particle flux in the wet season, associated with northwesterly winds (Fig. 5d), is likely a result of local pollution from the diesel generator (Ahlm et al., 2009) located within the research station (Fig. 1).

3.4.4 Diurnal cycles of the vertical particle flux

In this section, median diurnal cycles of the particle flux in the two seasons are investigated. Main focus is on the dry season particle flux and it is compared with the wet season particle flux. In order to exclude any possible impact from

the diesel generator and the house on the particle fluxes at K34, time periods with mean wind directions between 310 and 20 degrees have been excluded in the calculations of these diurnal cycles. In addition, time periods of rainfall have been ignored to simplify interpretation of the fluxes.

Figure 6 shows median diurnal cycles of the vertical particle flux in the dry and wet season. These fluxes have been calculated and de-trended over periods of 30 min. The particle flux is in general small at nighttime but larger in daytime when the turbulence intensity is much higher (Fig. 2i). In daytime, the median particle flux points downward both in the dry and wet season, indicating net deposition. The daytime deposition flux is significantly larger in the dry season than in the wet season. A larger deposition flux in the dry season is of course expected since anthropogenic impact on the aerosol population is significantly higher in the dry season, even though also the wet season particle flux contains some influence from anthropogenic sources.

The maximum deposition flux occurs in early afternoon and is $\sim 1.2 \times 10^6$ particles $\text{m}^{-2} \text{s}^{-1}$ in the dry season and $\sim 0.5 \times 10^6$ particles $\text{m}^{-2} \text{s}^{-1}$ in the wet season (Fig. 6). An approximate impact of these deposition fluxes on the particle concentration for each season can be estimated by using the median aerosol number concentration in Tables 1–2 for each season and assuming a maximum daytime mixed layer depth of 1100 m in the dry season and 1000 m in the wet season (Fisch et al., 2004). Then the deposition fluxes on average decrease the particle concentration around noon with 4.3 particles per cm^3 per hour in the dry season and 1.8 particles per

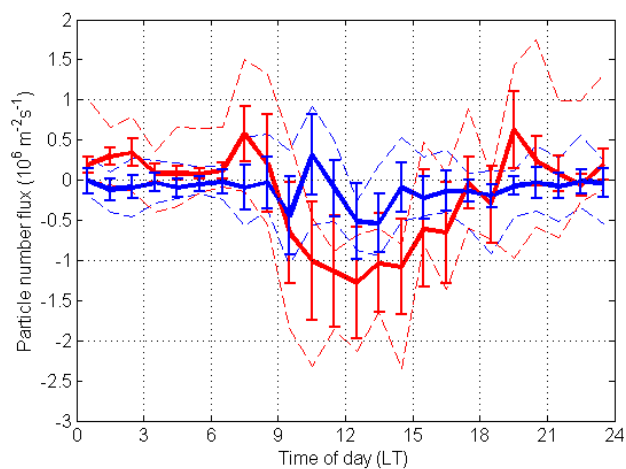


Fig. 6. Median diurnal cycles of particle flux (solid lines) in the dry season (red) and wet season (blue). Dashed lines are 25 and 75 percentiles. Error bars represent median random uncertainty.

cm^3 per hour in the wet season. The percentage loss is rather similar in the two seasons. About 0.3% of the total particle population is deposited per hour in early afternoon.

3.4.5 Upward particle fluxes

The wet season particle flux was analyzed in detail in Ahlm et al. (2009). There it was concluded that the upward flux peak between 10:00 and 11:00 LT most likely is a result of entrainment of cleaner air from above during fast mixed layer growth after the nocturnal inversion has been defeated. The dry season curve, however, has a quite large upward flux peak between 06:00 and 09:00 LT. At this time, the nocturnal inversion has not been defeated according to the discussion of the diurnal cycle of water vapor concentration in Sect. 3.2. This means that the upward flux peak is not likely a result of entrainment fluxes. The mixed layer is still thin this early in the morning which means that the associated turbulent time scales are short. Therefore fluxes calculated over 30 min (Fig. 6) are associated with large uncertainties. However, the median upward flux peak is apparent also when shorter time scales (10 and 3 min) are used for calculating the dry season flux (Fig. 7), and therefore these early morning upward particle fluxes seem reliable.

It is interesting to compare the median dry season diurnal cycle of the particle flux (Fig. 6) with the dry season diurnal cycle of CO_2 flux in Fig. 3. Obviously the peaks of the morning upward flux of particles and CO_2 occur at the same time. The peak in upward CO_2 flux in the morning was explained by release of CO_2 that has been stored within the canopy during the night, when conditions become more turbulent in the morning (Sect. 3.3). It is possible that also particles are being emitted from the forest throughout the whole night but stay confined within the canopy until turbulence starts increas-

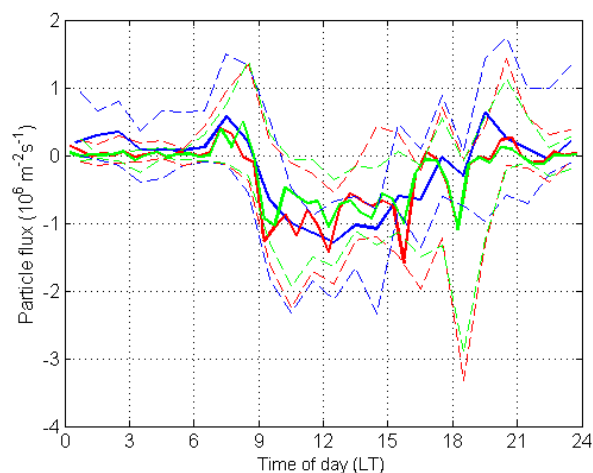


Fig. 7. Median diurnal cycles of the dry season aerosol number flux rotated and de-trended over 30 min (blue), 10 min (red) and 3 min (green). Dashed lines represent 25 and 75 percentiles.

ing after sunrise, which mixes up these particles so an upward flux appears at the altitude where the measurements are made, at the top of the tower K34. These dry season emission fluxes are not likely a result of local pollution, since the wind sector associated with advection from the diesel generator and the house have been excluded when calculating the diurnal cycles. Instead these upward fluxes actually might be a result of emission of natural biogenic particles from the forest.

In the case of CO_2 , it is very clear that the morning peak in upward flux is due to emission, because the CO_2 concentration peaks at the same time. However, the median diurnal cycle of particle concentration (Fig. 8) shows a different behavior than the diurnal cycle of CO_2 concentration. From midnight and until morning, the particle concentration decreases. The particle concentration actually continues its decreasing trend from the night when the upward particle flux appears in the morning. However, an emission source of $0.5 \times 10^6 \text{ particles m}^{-2} \text{ s}^{-1}$, like the early morning median upward flux in Fig. 6, active during one hour would only increase the particle concentration with 18 particles per cm^3 in a $\sim 100 \text{ m}$ thick boundary layer, which is only a little more than a one percent increase in particle concentration. The particle concentration in Fig. 8 shows a decreasing trend from midnight until 10:00 LT and the relatively small gain of particles from the emission flux in the morning is insignificant compared to the overall negative trend in concentration. Therefore, particle emission from the forest is still a possible explanation for the morning upward flux, even though there is no peak in particle concentration at the same time.

The median diurnal cycle of the dry season particle flux in Fig. 6 shows dominating upward fluxes also in the evening and throughout the night, particularly clear between 19:00 and 22:00 LT. The fact that the upward fluxes appear at

nighttime and in early morning does not necessarily mean that the possible emission source would be lower in daytime than at nighttime. Particles emitted at nighttime may be stored in the canopy layer which is decoupled from the atmosphere in stable conditions. Artaxo and Hansson (1995) and Guyon et al. (2003a, b), observed an increase in phosphorus concentration during nighttime at the lower part of the canopy, and they attributed this enhancement to nighttime biogenic emissions of particles containing phosphorus. Hence, the upward flux in the early morning would then be the flux of approximately all particles that have been emitted and stored under the canopy throughout the night. In daytime, when conditions are more turbulent, an emission of the same magnitude would generate upward fluxes that are more continuous and these emission fluxes would drown in the large daytime deposition flux.

Figure 9 shows an example of these early morning upward fluxes of particles and CO₂ and some related parameters. The figure shows how the parameters vary between 05:00 and 11:00 LT on 11 August. Both the night and following morning up until 11:00 LT were free from rainfall, and winds were blowing from the east, meaning that there was no influence from the research station. After a night of particle and CO₂ fluxes close to zero, upward fluxes appear shortly after 07:00 LT (Fig. 9a). These upward fluxes appear at approximately the same time as the friction velocity starts to increase (Fig. 9b). Interestingly, the heat flux (Fig. 9b) is negative at the same time as the upward fluxes appear. This means that the stratification is still stable and that the nocturnal inversion has not been dissipated yet. Furthermore, the concentration of water vapor (Fig. 9c) and CO₂ (Fig. 9d) increases when the upward fluxes appear. In fact, also the particle concentration increases from 07:00 LT to slightly after 08:00 LT at the same time as the upward particle flux increases. Therefore the upward fluxes cannot likely be explained by dilution from above by entrainment. In Fig. 9, it seems as the nocturnal inversion is dissipated around 09:00 LT. At this time, the sensible heat flux becomes positive and the water vapor concentration starts to decrease.

If the upward fluxes are associated with emission from the rain forest, it is not likely that the emitted particles are secondary aerosol particles. Numbers of nucleation mode particles are low in the Amazon boundary layer. Whereas in other continental locations 3-nm particles are regularly observed at near-surface measurement sites, in the Amazon Basin the smallest particles typically have sizes of 10 to 20 nm (Martin et al., 2010). This has led to the hypothesis that new particle formation may occur at higher altitudes, which means that the occasionally observed 10–20 nm particles in the surface layer have not likely been formed close to the surface. Hence, a source of primary biogenic aerosol particles is a more likely explanation for the observed upward fluxes in the morning. To investigate this further one would have to measure particle concentration in the canopy layer in parallel with particle concentrations above the canopy, and inves-

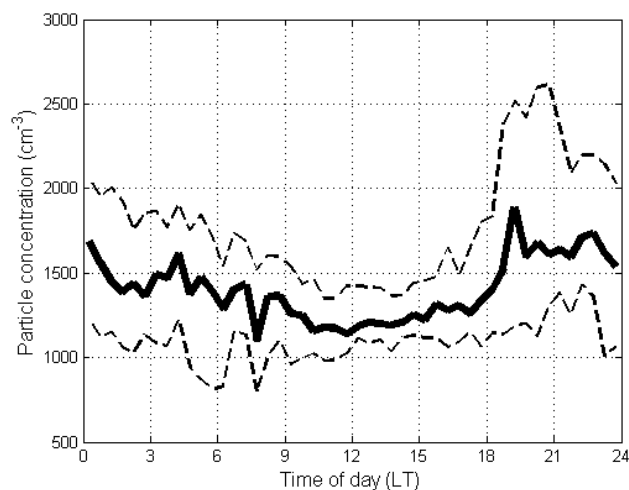


Fig. 8. Median diurnal cycle of the dry season aerosol number concentration (solid line) with 25 and 75 percentiles (dashed lines).

tigate whether a higher aerosol number concentration is built up in the canopy layer throughout the night simultaneously with the observed higher phosphorus concentration.

3.4.6 Particle transfer velocities

There is no perfect way of estimating the particle deposition velocity when both emission and deposition contribute to the vertical net flux. Nor is it always possible to know whether emission contributes to the net flux, since emission could make a contribution even when the net flux points downward.

In this study we define the particle transfer velocity as

$$v_t = -\frac{F}{N} \quad (2)$$

where F is the particle number flux and N is the particle number concentration. Positive values on v_t represents a net downward flux. To estimate the average deposition velocity by calculating an average value of v_t over a whole data set, according to Eq. (2), will underestimate the deposition velocity if processes like entrainment (Nilsson et al., 2001; Ahlm et al., 2009) or surface emission temporarily produce net upward fluxes. However, a dominating part of the net upward fluxes in this study are likely due to random errors, why these should not be excluded.

Figure 10 shows the median diurnal cycles of v_t both in the dry and wet season for fluxes calculated and de-trended over time scales of 30 min. v_t is low at nighttime but higher in daytime when conditions are more turbulent. The daytime particle transfer velocities have rather similar values in the two seasons. At nighttime, the transfer velocities have different signs in the two seasons as a result of net downward fluxes at nighttime in the wet season and net upward fluxes at nighttime in the dry season.

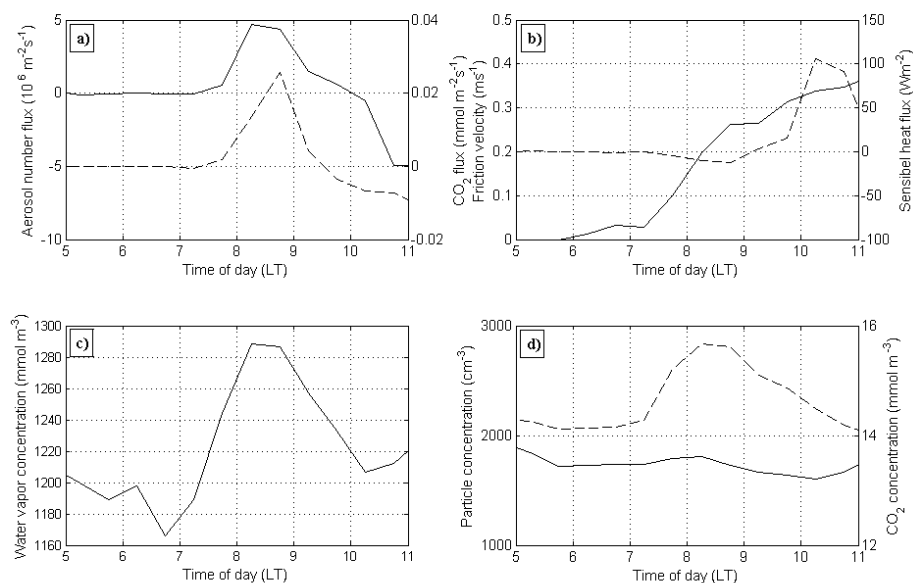


Fig. 9. (a) Aerosol number flux (solid line) and CO₂ flux (dashed line), (b) friction velocity (solid line) and sensible heat flux (dashed line), (c) water vapor concentration, and (d) particle concentration (solid line) and CO₂ concentration (dashed line). (a–d) show the variation of the parameters between 05:00 and 11:00 LT on 11 August.

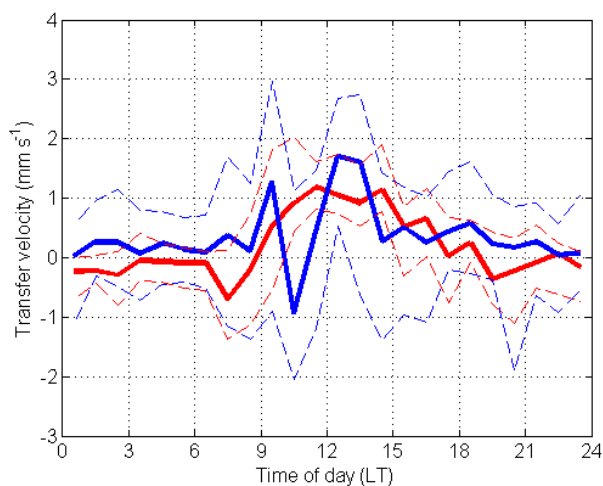


Fig. 10. Median diurnal cycles of v_t in the dry (red) and wet (blue) season. Dashed lines represent 25 and 75 percentiles.

In general, transfer velocities are low here compared to several dry deposition studies over boreal forests (Ruijgrok et al., 1997; Buzorius et al., 2000; Gaman et al., 2004). Dominance of accumulation mode particles and low numbers of nucleation mode particles in the Amazon boundary layer, both in the dry and wet season, are likely important factors for these low values on v_t . Pryor et al. (2007) measured dry deposition velocities with a relaxed eddy accumulation (REA) system in the particle diameter range 10–100 nm in a deciduous forest at Sorø in Denmark and in a pine forest at Hyytiälä in Finland. They observed decreasing deposition velocity with increasing particle geometric mean diameter in

this diameter interval. For geometric mean diameters above 50 nm, the median deposition velocity was below 2 mm s^{-1} . Since the aerosol number size distribution in the Amazon boundary layer is dominated by particle diameters where the deposition velocity (as function of particle diameter) is at its minimum, low particle transfer velocities are logical in the Amazon basin. Another important reason for the low transfer velocities is of course the low wind speeds in the tropics compared to the midlatitudes.

When considering the fact that wet deposition is a very important deposition process over tropical rain forests (as a result of the high rain amounts) and adding the low particle transfer velocities found in this study, it can be stated that the relative contribution of dry deposition to total deposition of particles is much lower in the continental tropics than in the continental midlatitudes. In this way, the continental tropics resemble many marine environments.

3.4.7 Transfer velocity dependence on friction velocity

In both the dry and wet season, downward fluxes strongly dominate in the afternoon (Fig. 6). At this time the mixed layer is well developed, which means that there is not much disturbance from entrainment. Therefore, the transfer velocities in the afternoon are likely good estimations of the deposition velocity. Figure 11 shows how the particle transfer velocity depends on friction velocity when only particle fluxes measured between 12:00 and 17:00 LT are included. Obviously, the transfer velocity increases linearly with increasing friction velocity in both seasons. The transfer velocities seem to have very similar values in the two seasons.

Following relations describe the linear fits shown in Fig. 11 (in which the R^2 values have been calculated for the binned data):

$$v_t = (2.63u_* + 0.04) \times 10^{-3} (R^2 = 0.84) \quad (3)$$

in the dry season, and

$$v_t = (2.78u_* - 0.04) \times 10^{-3} (R^2 = 0.61) \quad (4)$$

in the wet season.

Most studies of dry deposition for particles have shown that the minimum deposition velocity is located at diameters around 0.1–0.3 μm (Zhang and Vet, 2006). For lower particle sizes, Brownian diffusion becomes more efficient and for larger sizes interception and impaction become increasingly important (Slinn, 1982). As been discussed in earlier sections, the percentage of accumulation mode particles within the Amazon boundary layer increases with a following percentage decrease of Aitken mode particles during transition from wet season to dry season (Table 3). A change towards a higher percentage of particles in the accumulation mode in the dry season will reduce the efficiency of dry deposition since the efficiency of Brownian diffusion decreases with increasing particle size. However, the very similar values in dry and wet season values on v_t (Fig. 11) indicate that the change in size distribution between the wet and dry season is not enough to have a significant impact on the overall particle deposition velocity. There is of course the possibility that particle emission from the rain forest cancels part of the deposition. An emission flux of a certain magnitude would cancel a larger fraction of the deposition flux in the wet season than in the dry season, since the deposition flux is much smaller in the wet season due to lower particle concentrations.

Another factor that might have an influence is the seasonal variations in leaf area index (LAI). The LAI has been observed to increase during the dry season with as much as 25% from the annual mean (Myneni et al., 2007). A larger LAI means more area for particles to deposit on. Hence, a larger dry season LAI could increase the dry season particle deposition velocities, thereby also counteracting the effect of higher dry season percentage of accumulation mode particles. The difference in impact of particle rebound in dry and wet conditions, respectively, is probably of less importance since particle bounce off primarily affects coarse particles, which are very low in numbers, and therefore do not have a large influence on particle fluxes measured with CPC.

3.4.8 Aerodynamic resistance and surface transfer velocity

The surface transfer velocity can be defined as

$$v_{ts} = \frac{1}{\frac{1}{v_t} - r_a} \quad (5)$$

where r_a is the aerodynamic resistance.

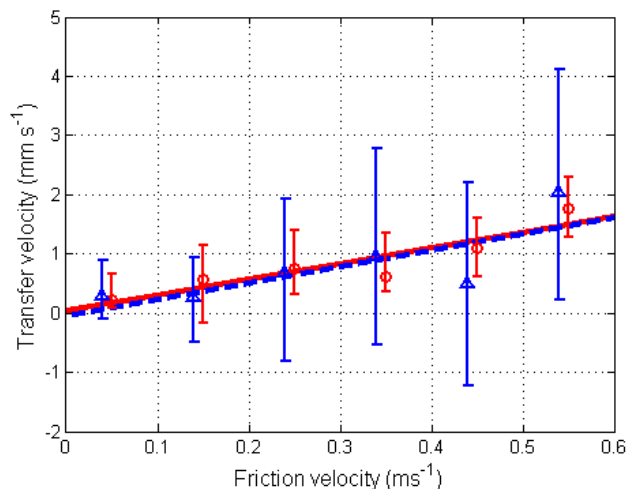


Fig. 11. Median values of v_t over constant friction velocity intervals in the dry season (red circles) and wet season (blue triangles), and linearly fitted curves to the dry season (solid red line) and wet season (dashed blue line) data. The figure represents data collected between 12:00 and 17:00 LT. The vertical bars represent 25 and 75 percentiles. The values of the medians and the percentiles have in each bin been calculated over a minimum of 14 half-hour values.

In this study, we have made a rough estimate of the aerodynamic resistance by using relations given in Seinfeld and Pandis et al. (1998) with assumed values on the roughness length and the displacement height as 1.8 m and 25.8 m, respectively (Harris et al., 2004). r_a is high at nighttime and low in daytime (Fig. 12a). Figure 12b shows median diurnal cycles of v_t and v_{ts} . Obviously the difference between v_t and v_{ts} is very small. This can also be realized only by studying Eq. (5) for a reasonable range of values of v_t and r_a in this study. Equations (3–4) describing the transfer velocity as functions of friction velocity were not converted to surface transfer velocity. It also seems preferable not to convert v_t to v_{ts} when not necessary, since the estimations of r_a in this study are only rough estimations, however, probably accurate enough to state that $v_t \approx v_{ts}$. Therefore, we suggest that Eqs. (3–4) can be used for estimating the surface transfer velocity in models.

4 Summary and conclusions

Aerosol number fluxes and CO_2 fluxes were measured with the eddy covariance method over the Amazon rain forest in 2008 in both the dry and wet season. The measurements were performed at the top of the 53 m high tower K34 in the Cuieiras Reserve, Manaus, Brazil. Aerosol number fluxes measured during the dry season, when the impact from biomass burning is higher, are compared with fluxes measured in the much cleaner conditions prevailing in the wet season. The key results and main conclusions are:

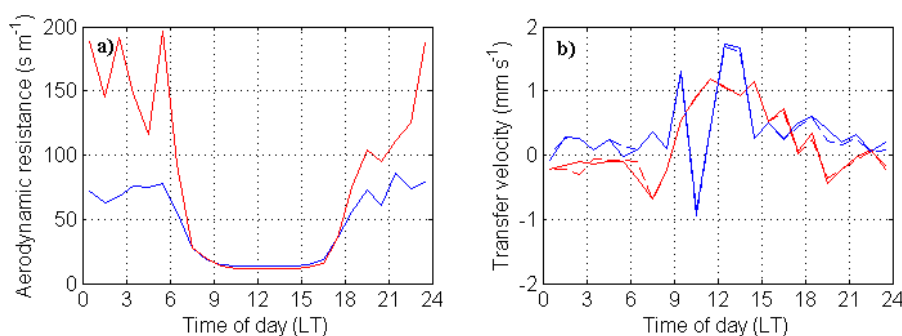


Fig. 12. (a) Rough estimation of the median diurnal cycle of aerodynamic resistance in the dry season (solid red line) and wet season (dashed blue line). (b) v_{ts} (solid lines) and v_t (dashed lines) in the dry (red) and wet season (blue).

- The median aerosol number concentration was $1352\ cm^{-3}$ in the dry season and $466\ cm^{-3}$ in the wet season.
- Particle transfer velocities peak around noon or in early afternoon at values of $1\text{--}2\ mm\ s^{-1}$ both in the dry and wet season. The daytime particle transfer velocities generally have very similar values in the two seasons.
- The particle transfer velocity v_t increases linearly with increasing friction velocity in both seasons. The relations are described by $v_t = (2.63u_* + 0.04) \times 10^{-3}$ in the dry season and $v_t = (2.78u_* - 0.04) \times 10^{-3}$ in the wet season.
- Particle transfer velocities are low in this study in comparison to measurements made over boreal forests. This is likely due to dominance of accumulation mode particles and low numbers of nucleation mode particles in the Amazon boundary layer, both in the dry and wet season. Another important reason is the low wind speeds in the tropics compared to the midlatitudes. When considering the fact that wet deposition is a very important deposition process over tropical rain forests and adding the low particle transfer velocities found in this study, it can be stated that the relative contribution of dry deposition to total deposition of particles is much lower over tropical rain forests than over boreal forests, and instead comparable to many marine regions.
- Net particle deposition prevails in daytime both in the dry and wet season. This deposition flux is much larger in the dry season than in the wet season. The much larger deposition flux in the dry season is a result of the higher dry season aerosol number concentration.
- In the dry season, nocturnal particle fluxes behave very similar to nocturnal CO_2 fluxes. Particle fluxes are very low in magnitude throughout the night but after sunrise upward particle fluxes appear. These appear before the nocturnal inversion has been defeated and are

therefore not likely a result of entrainment. Nor does local pollution seem to be a likely explanation for these upward fluxes, since associated wind sectors have been excluded. Emission of natural biogenic particles from the forest, however, is a possible explanation. The upward flux appears at the same time as the CO_2 emission flux. It is possible that particles are emitted throughout the night but stay within the canopy, which is decoupled from the atmosphere above, until turbulence mixes them up in the morning, similarly to what is observed for CO_2 . It is also possible that they are emitted throughout the day, but then are masked by the larger deposition flux.

Hence, this study has shown that particle transfer velocities are very similar in the dry and wet season, which indicates that the change in aerosol number size distribution between the two seasons is not enough to result in a significant change in average deposition velocity. It would be interesting to make the same dry/wet season comparison in the southern part of the Amazon rain forest where the impact from biomass burning on the dry season aerosol population is much larger.

Upward particle fluxes with the magnitude of $0.5 \times 10^6\ m^{-2}\ s^{-1}$, like the observed morning upward flux in this study, would only increase the particle concentration with 18 particles per cm^3 and hour in a 100 m thick boundary layer. However, since nocturnal respiration is known to be underestimated by eddy covariance measurements, it is likely that also nocturnal particle emission is underestimated.

Acknowledgements. We would like to thank the National Institute for Research in the Amazon (INPA) and the LBA infrastructure team for all help and support during this campaign. Paulo Artaxo acknowledges financial support from the CNPq/MCT Millennium Institute Program and FAPESP. We thank Paulo Henrique Oliveira (in memoriam) for support during the whole sampling campaign.

Swedish participation was provided by Swedish International Development Cooperation Agency (Sida/SAREC), the Swedish Research Council (VR) and from the 50th birthday of King Carl XVI

Gustav Foundation for Science Technology and Environment. We also acknowledge Leif Bäcklin and Kai Rosman for technical assistance, and the Bert Bolin centre for climate research financial support. Finally, we thank Peter Tunved and Luciana V. Rizzo for assistance in data analysis, and Üllar Rannik for good advices.

Edited by: I. Trebs

References

- Ackerman, A. S., Toon, O. B., Stevens, D. E., Heymsfield, A. J., Ramanathan, V., and Welton, E. J.: Reduction of Tropical Cloudiness by Soot, *Science*, 288, 1042–1047, 2000.
- Ahlm, L., Nilsson, E. D., Krejci, R., Mårtensson, E. M., Vogt, M., and Artaxo, P.: Aerosol number fluxes over the Amazon rain forest during the wet season, *Atmos. Chem. Phys.*, 9, 9381–9400, 2009, <http://www.atmos-chem-phys.net/9/9381/2009/>.
- Andreae, M. O., Browell, E. V., Garstang, M., Gregory, G. L., Harriss, R. C., Hill, G. F., Jacob, D. J., Pereira, M. C., Sachse, G. W., Setzer, A. W., Silva Dias, P. L., Talbot, R. W., Torres, A. L., and Wofsy, S. C.: Biomass-burning emissions and associated haze layers over Amazonia, *J. Geophys. Res.*, 93, 1509–1527, 1988.
- Andreae, M. O. and Merlet, P.: Emission of trace gases and aerosols from biomass burning, *Global Biogeochem. Cy.*, 15(4), 955–966, 2001.
- Andreae, M. O., Artaxo, P., Fischer, H., Freitas, S. R., Gregoire, J. M., Hansel, A., Hoor P., Kormann, R., Krejci, R., Lange, L., Lelieveld, J., Lindinger, W., Longo, K., Peters, W., de Reus, M., Scheeren, B., Dias, M. A. F. S., Ström, J., van Velthoven, P. F. J., and Williams, J.: Transport of biomass burning smoke to the upper troposphere by deep convection in the equatorial region, *Geophys. Res. Lett.*, 28(6), 951–954, 2001.
- Andreae, M. O., Rosenfeld, D., Artaxo, P., Costa, A. A., Frank, G. P., Longo, K. M., and Silva-Dias, M. A. F.: Smoking rain clouds over the Amazon, *Science*, 303, 1337–1342, 2004.
- Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, *Atmos. Chem. Phys.*, 6, 3131–3148, 2006, <http://www.atmos-chem-phys.net/6/3131/2006/>.
- Araújo, A. C., Kruijt, B., Nobre, A. D., Dolman, A. J., Maarten, J. W., Moors, E. J., and Souza, J. S.: Nocturnal accumulation of CO₂ underneath a tropical forest canopy along a topographical gradient, *Ecol. Appl.*, 18(6), 1406–1419, 2008.
- Artaxo, P., Gerab, F., Yamasoe, M. A., and Martins, J. V.: Fine Mode Aerosol Composition in Three Long Term Atmospheric Monitoring Sampling Stations in the Amazon Basin, *J. Geophys. Res.*, 99(D11), 22857–22868, 1994.
- Artaxo, P. and Hansson, H.-C.: Size distribution of biogenic aerosol particles from the Amazon basin, *Atmos. Environ.*, 29, 393–402, doi:10.1016/1352-2310(94)00178-N, 1995.
- Artaxo, P., Fernandes, E. T., Martins, J. V., Yamasoe, M. A., Hobbs, P. V., Maenhaut, W., Longo, K. M., and Castanho, A.: Large-scale aerosol source apportionment in Amazonia, *J. Geophys. Res.*, 103, 31837–31847, 1998.
- Artaxo, P., Martins, J. V., Yamasoe, M. A., Procópio, A. S., Pauliquevis, T. M., Andreae, M. O., Guyon, P., Gatti, L. V., and Leal, A. M. C.: Physical and chemical properties of aerosols in the wet and dry season in Rondonia, Amazonia, *J. Geophys. Res.*, 107, 8081, doi:10.1029/2001JD000666, 2002.
- Aubinet, M., Heinesch, B., and Yernaux, M.: Horizontal and vertical CO₂ advection in a sloping forest, *Bound. Lay. Meteorol.*, 108(3), 397–417, 2003.
- Bowman, D. M. J. S., Balch, J. K., Artaxo, P., Bond, W. J., Carlson, J. M., Cochrane, M. A., D'Antonio, C. M., DeFries, R. S., Doyle, J. C., Harrison, S. P., Johnston, F. H., Keeley, J. E., Krawchuk, M. A., Kull, C. A., Marston, J. B., Moritz, M. A., Prentice, I. C., Roos, C. I., Scott, A. C., Swetnam, T. W., van der Werf, G. R., and Pyne, S. J.: Fire in the Earth System, *Science*, 324, 481–484, doi:10.1126/science.1163886, 2009.
- Buzorius, G., Rannik, Ü, Mäkelä, J. M., Keronen, P., Vesala, T., and Kulmala, M.: Vertical aerosol fluxes measured by the eddy covariance method and deposition of nucleation mode particles above a Scots pine forest in southern Finland, *J. Geophys. Res.*, 105, 19905–19916, 2000.
- Campos, J. G., Acevedo, O. C., Tóta, J., and Manzi, A. O.: On the temporal scale of the turbulent exchange of carbon dioxide and energy above a tropical rain forest in Amazonia, *J. Geophys. Res.*, 114, D08124, doi:10.1029/2008JD011240, 2009.
- Doebelin, E. O.: *Measurement Systems*, McGraw-Hill, New York, 1990.
- Elbert, W., Taylor, P. E., Andreae, M. O., and Pöschl, U.: Contribution of fungi to primary biogenic aerosols in the atmosphere: wet and dry discharged spores, carbohydrates, and inorganic ions, *Atmos. Chem. Phys.*, 7, 4569–4588, 2007, <http://www.atmos-chem-phys.net/7/4569/2007/>.
- Finnigan, J., Clement, R., Malhi, Y., Leuning, R., and Cleugh, H. A.: A re-evaluation of long-term flux measurement techniques. Part I. Averaging and coordinate rotation, *Bound. Lay. Meteorol.*, 107, 1–48, 2003.
- Finnigan, J.: An introduction to flux measurements in difficult conditions, *Ecol. Appl.*, 18(6), 1340–1350, 2008.
- Fish, B. R.: Electrical generation of natural aerosols from vegetation, *Science*, 175, 1239–1240, 1972.
- Fisch, G., Tóta, J., Machado, L. A. T., Dias, M., Lyra, R. F. D., Nobre, C. A., Dolman, A. J., and Gash, J. H. C.: The convective boundary layer over pasture and forest in Amazonia, *Theor. Appl. Clim.*, 78, 47–59, 2004.
- Foken, T. and Wichura, B.: Tools for quality assessment of surface-based flux measurements, *Agr. Forest Meteorol.*, 78, 83–105, 1996.
- Gaman, A., Rannik, Ü, Aalto, P., Pohja, T., Siivola, E., Kulmala, M. and Vesala T.: Relaxed eddy accumulation system for size resolved aerosol flux measurements, *J. Atmos. Ocean. Tech.*, 21, 933–943, 2004.
- Garstang, M., Ulanski, S., Greco, S., Scala, J., Swap, R., Fitzjarrald, D., Browell, E., Shipman, M., Connors, V., Harriss, R., and Talbot, R.: The Amazon Boundary-Layer Experiment (ABLE 2B): a meteorological perspective, *B. Am. Meteorol. Soc.*, 71, 19–31, 1990.
- Goulden, M. L., Munger, J. W., Fan, S. M., Daube, B. C., and Wofsy, S. C.: Measurements of carbon sequestration by long-term eddy covariance: Methods and a critical evaluation of accuracy, *Glob. Change Biol.*, 2, 169–182, 1996.
- Graham, B., Guyon, P., Maenhaut, W., Taylor, P. E., Ebert, M., Matthias-Maser, S., Mayol-Bracero, O. L., Godoi, R., Artaxo, P., Meixner, F. X., Moura, M. A., Rocha, C. H., Grieken, R. V.,

- Glovsky, M., Flagan, R., and Andreae, M. O.: Composition and diurnal variability of the natural Amazonian aerosol. *J. Geophys. Res.*, 108(D24), 4765, doi:10.1029/2003JD004049, 2003.
- Gunthe, S. S., King, S. M., Rose, D., Chen, Q., Roldin, P., Farmer, D. K., Jimenez, J. L., Artaxo, P., Andreae, M. O., Martin, S. T., and Pöschl, U.: Cloud condensation nuclei in pristine tropical rainforest air of Amazonia: size-resolved measurements and modeling of atmospheric aerosol composition and CCN activity, *Atmos. Chem. Phys.*, 9, 7551–7575, 2009, <http://www.atmos-chem-phys.net/9/7551/2009/>.
- Guyon, P., Graham, B., Beck, J., Boucher, O., Gerasopoulos, E., Mayol-Bracero, O. L., Roberts, G. C., Artaxo, P., and Andreae, M. O.: Physical properties and concentration of aerosol particles over the Amazon tropical forest during background and biomass burning conditions, *Atmos. Chem. Phys.*, 3, 951–967, 2003a, <http://www.atmos-chem-phys.net/3/951/2003/>.
- Guyon, P., Graham, B., Roberts, G. C., Mayol-Bracero, O. L., Maenhaut, W., Artaxo, P., and Andreae, M. O.: In-canopy gradients, composition, sources, and optical properties of aerosol over the Amazon forest, *J. Geophys. Res.*, 108(D18), 4591, doi:10.1029/2003JD003465, 2003b.
- Harris, P. P., Hunting, C., Cox, P. M., Gash, J. H. C., and Malhi, Y.: Effect of soil moisture on canopy conductance of Amazonian rainforest, *Agr. Forest Meteorol.*, 122, 215–227, 2004.
- Hoppel, W. A., Frick, G. M., and Fitzgerald, J. W.: A cloud chamber study of the effect that non precipitating water clouds have on aerosol size distribution, *Aerosol Sci. Tech.*, 20, 1–30, 1994.
- Horst, T. W.: A simple formulation for attenuation of eddy fluxes measured with first-order-response scalar sensors, *Bound. Lay. Meteorol.*, 82, 219–233, 1997.
- Jones, A. M. and Harrison, R. M.: The effects of meteorological factors on atmospheric bioaerosol concentrations – a review, *Sci. Total Environ.*, 326, 151–180, 2004.
- Järvi, L., Rannik, Ü., Mammarella, I., Sogachev, A., Aalto, P. P., Keronen, P., Siivola, E., Kulmala, M., and Vesala, T.: Annual particle flux observations over a heterogeneous urban area, *Atmos. Chem. Phys.*, 9, 7847–7856, 2009, <http://www.atmos-chem-phys.net/9/7847/2009/>.
- Koren I., Martins, J. V., Remer, L. A., and Afargan, H.: Smoke invigoration versus inhibition of clouds over the Amazon, *Science*, 321, 946–949, 2008.
- Kruijt, B., Malhi, Y., Lloyd, J., Nobre, A. D., Miranda, A. C., Pereira, M. G. P., Culf, A., and Grace, J.: Turbulence above and within two Amazon rainforest canopies, *Bound. Lay. Meteorol.*, 94, 297–311, 2000.
- Lenshow, D. H. and Raupach, M. R.: The attenuation of fluctuations in scalar concentrations through sampling tubes, *J. Geophys. Res.*, 96, 5259–5268, 1991.
- Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, *Atmos. Chem. Phys.*, 7, 3425–3446, 2007, <http://www.atmos-chem-phys.net/7/3425/2007/>.
- Malhi, Y., Nobre, A. D., Grace, J., Kruijt, B., Pereira, M. G. P., Culf, A., and Scott, S.: Carbon dioxide transfer over a central Amazonian rain forest, *J. Geophys. Res.*, 103, 31593–31612, 1998.
- Marcolla, B., Cescatti, A., Montagnani, L., Manca, G., Kerschbaumer, G., and Minerbi, S.: Importance of advection in the atmospheric CO₂ exchanges of an alpine forest, *Agr. Forest Meteorol.*, 130, 193–206, 2005.
- Martin, S. T., Andreae, M. O., Artaxo, P., Baumgardner, D., Chen, Q., Goldstein, A. H., Guenther, A., Heald, C. L., Mayol-Bracero, O. L., McMurry, P. H., Pauliquevis, T., Pöschl, U., Prather, K. A., Roberts, G. C., Saleska, S. R., Silva Dias, M. A., Spracklen, D. V., Swietlicki, E., and Trebs, I.: Sources and Properties of Amazonian Aerosol Particles, *Rev. Geophys.*, in press, 2010.
- Myneni, R. B., Yang, W., Nemani R. R., Huete, A. R., Dickinson, R. E., Knyazikhina, Y., Didan, K., Fue, R., Juárez, R. I. N., Saatchi, S. S., Hashimoto, H., Ichii, K., Shabanov, N. V., Tan, B., Ratana, P., Privette, J. L., Morisette, J. T., Vermotek, E. F., Roy, D. P., Wolfe, R. E., Friedl, M. A., Running, S. W., Votava, P., El-Saleous, N., Devadiga, S., Su, Y., and Salomonson, V.: Large seasonal swings in leaf area of Amazon rain forest, *P. Natl. Acad. Sci. USA*, 104, 4820–4823, 2007.
- Nemeruyk, G. E.: Migration of salts into the atmosphere during transpiration, *Soviet Plant Physiol.* 17, 560–566, 1970.
- Nilsson, E. D., Rannik, Ü., Buzorius, G., Kulmala, M., and O’Dowd, C.: Effects of the continental boundary layer evolution, convection, turbulence and entrainment on aerosol formation, *Tellus*, 53B, 441–461, 2001.
- Oliveira, P. H. F., Artaxo, P., Pires, C., Lucca, S., Procopio, A., Holben, B., Schafer, J., Cardoso, L. F., Wofsy, S. C., and Rocha, H. R.: The effect of biomass burning aerosols and clouds on the CO₂ flux in Amazonia, *Tellus*, 59B, 338–349, 2007.
- Pryor, S. C., Larsen, S. E., Sørensen, L. L., Barthelmie, R. J., Grönholm, T., Kulmala, M., Launiainen, S., Rannik, Ü., and Vesala, T.: Particle fluxes over forests: Analyses of flux methods and functional dependencies, *J. Geophys. Res.*, 112, D07205, doi:10.1029/2006JD008066, 2007.
- Rannik, Ü., Mammarella, I., Aalto, P., Keronen, P., Vesala, T., and Kulmala, M.: Long-term aerosol particle flux observations part : Uncertainties and time-average statistics, *Atmos. Environ.*, 43, 3431–3439, 2009.
- Reid, J. S., Koppmann, R., Eck, T. F., and Eleuterio, D. P.: A review of biomass burning emissions part II: intensive physical properties of biomass burning particles, *Atmos. Chem. Phys.*, 5, 799–825, 2005, <http://www.atmos-chem-phys.net/5/799/2005/>.
- Rissler, J., Swietlicki, E., Zhou, J., Roberts, G., Andreae, M. O., Gatti, L. V., and Artaxo, P.: Physical properties of the sub-micrometer aerosol over the Amazon rain forest during the wet-to-dry season transition - comparison of modeled and measured CCN concentrations, *Atmos. Chem. Phys.*, 4, 2119–2143, 2004, <http://www.atmos-chem-phys.net/4/2119/2004/>.
- Rissler, J., Vestin, A., Swietlicki, E., Fisch, G., Zhou, J., Artaxo, P., and Andreae, M. O.: Size distribution and hygroscopic properties of aerosol particles from dry-season biomass burning in Amazonia, *Atmos. Chem. Phys.*, 6, 471–491, 2006, <http://www.atmos-chem-phys.net/6/471/2006/>.
- Rossman, A. Y.: Strategy for an all-taxa inventory of fungal biodiversity, in: *Biodiversity and Terrestrial Ecosystems*, Acad. Sinica Monograph Series, 14, 169–194, 1994.
- Ruijgrok, W., Tieben, H., and Eisinga, P.: The dry deposition of particles to a forest canopy: A comparison of model and experimental results, *Atmos. Environ.*, 31, 399–415, 1997.
- Schnell, R. C. and Vali, G.: Atmospheric ice nuclei from decomposing vegetation, *Nature*, 236, 163–165, 1973.
- Seinfeld, J. H. and Pandis, S. N.: *Atmospheric chemistry and*

- physics, John Wiley & Sons, 1998.
- Slinn, W. G. N.: Predictions for particle deposition to vegetative canopies, *Atmos. Environ.*, 16, 1785–1794, 1982.
- Stull, R. B.: An introduction to boundary layer meteorology, Kluwer Academic Publishers, The Netherlands, 1988.
- Thompson, W. A. R. (ed.): Black's medical dictionary, 33 ed. Adam and Charles Black, 1981.
- Wyngaard, J. C.: On surface-layer turbulence, in: Workshop on micrometeorology, edited by: Haugen, D. A., American Meteorological Society, Boston, Mass., 101–149, 1973.
- Zhang, L. and Vet, R.: A review of current knowledge concerning size-dependent aerosol removal, *China Particuology*, 4, 272–282, 2006.
- Zhou, J. C., Swietlicki, E., Hansson, H. C., and Artaxo, P.: Sub-micrometer aerosol particle size distribution and hygroscopic growth measured in the Amazon rain forest during the wet season, *J. Geophys. Res.*, 107, 8055, doi:10.1029/2000JD000203, 2002.