

1 Aerosol profiling with lidar in the Amazon Basin during the wet 2 and dry season

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6 [1] For the first time, multiwavelength polarization Raman lidar observations of optical
7 and microphysical particle properties over the Amazon Basin are presented. The fully
8 automated advanced Raman lidar was deployed 60 km north of Manaus, Brazil (2.5°S,
9 60°W) in the Amazon rain forest from January to November 2008. The measurements thus
10 cover both the wet season (Dec–June) and the dry or burning season (July–Nov). Two
11 cases studies of young and aged smoke plumes are discussed in terms of spectrally resolved
12 optical properties (355, 532, and 1064 nm) and further lidar products such as particle
13 effective radius and single-scattering albedo. These measurement examples confirm that
14 biomass burning aerosols show a broad spectrum of optical, microphysical, and chemical
15 properties. The statistical analysis of the entire measurement period revealed strong
16 differences between the pristine wet and the polluted dry season. African smoke and dust
17 advection frequently interrupt the pristine phases during the wet season. Compared to
18 pristine wet season conditions, the particle scattering coefficients in the lowermost 2 km of
19 the atmosphere were found to be enhanced, on average, by a factor of 4 during periods of
20 African aerosol intrusion and by a factor of 6 during the dry (burning) season. Under
21 pristine conditions, the particle extinction coefficients and optical depth for 532 nm
22 wavelength were frequently as low as 10–30 Mm⁻¹ and <0.05, respectively. During the
23 dry season, biomass burning smoke plumes reached to 3–5 km height and caused a mean
24 optical depth at 532 nm of 0.26. On average during that season, particle extinction
25 coefficients (532 nm) were of the order of 100 Mm⁻¹ in the main pollution layer (up to
26 2 km height). Ångström exponents were mainly between 1.0 and 1.5, and the majority of
27 the observed lidar ratios were between 50–80 sr.

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31 1. Introduction

32 [2] The Amazon Basin is the largest hydrological basin in
33 the world containing the largest extent of tropical rain forest
34 on Earth – the Amazon rain forest. The tropical rain forest
35 covers more than 5,000,000 square kilometers [*Nobre et al.*,
36 2004] and thus an area half as large as Europe or rather one
37 third of South America. Because of its size and its

pronounced hydrological cycle, the Amazon Basin is a key 38
region for the global climate. 39

[3] From field campaigns during the last three decades 40
(see review of *Martin et al.* [2010a]) it was concluded that 41
Amazonia is at times very clean and free of anthropogenic 42
influences in the wet season, while during the dry season 43
smoke from vegetation fires heavily influences the atmo- 44
spheric conditions. *Pöschl et al.* [2010] state that aerosol 45
conditions in Amazonia’s wet season “approach to those of 46
the pristine pre-industrial era”. Because of this strong con- 47
trast between the wet and the dry season, the Amazon Basin 48
is considered to be favorable to study the direct and indirect 49
aerosol effect on climate. 50

[4] Aerosol research in Amazonia was predominantly 51
performed during the dry season with focus on biomass- 52
burning aerosol (BBA) [*Kaufman et al.*, 1992; *Ward et al.*, 53
1992; *Kaufman et al.*, 1998; *Andreae et al.*, 2004]. During 54
several field campaigns microphysical and optical properties 55
of smoke aerosol were investigated mainly at the surface 56
with in situ instrumentation [*Artaxo et al.*, 1994, 2002; 57

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58 Guyon *et al.*, 2003] and by means of airborne measurements
 59 [Reid *et al.*, 1998; Reid and Hobbs, 1998; Chand *et al.*,
 60 2006]. Based on such BBA measurements, estimations of
 61 the direct aerosol effect were made [Ross *et al.*, 1998] and
 62 hypotheses were formulated concerning the aerosol semi-
 63 direct [Koren *et al.*, 2004] and indirect effect [Reid *et al.*,
 64 1999; Andreae *et al.*, 2004; Koren *et al.*, 2004; Rosenfeld
 65 *et al.*, 2008]. However, most of these campaigns were per-
 66 formed in the southern cerrado regions and thus may not be
 67 representative for the entire Amazon Basin.

68 [5] Aerosol research in the wet season was less frequent
 69 and focused on natural aerosol from the rain forest. Biogenic
 70 aerosol (primary organic and secondary organic aerosol)
 71 from the forest was identified as the dominant aerosol spe-
 72 cies during that season [Artaxo *et al.*, 1988; Martin *et al.*,
 73 2010b].

74 [6] Events of Saharan dust advection occasionally take
 75 place during the wet season and can significantly change the
 76 atmospheric aerosol conditions over the Amazon rain forest
 77 during that time [Talbot *et al.*, 1990; Artaxo *et al.*, 1990;
 78 Swap *et al.*, 1992; Formenti *et al.*, 2001; Ben-Ami *et al.*,
 79 2010]. However, recently it was found that also BBA from
 80 African vegetation fires reaches the Amazon Basin together
 81 with Saharan dust and significantly disturbs the clean back-
 82 ground conditions [Kaufman *et al.*, 2005; Ansmann *et al.*,
 83 2009; Baars *et al.*, 2011]. BBA then frequently dominates
 84 the optical aerosol properties.

85 [7] As a consequence of the low natural aerosol con-
 86 centrations, the impact of anthropogenic aerosol on rainfall
 87 production (aerosol indirect effect) may have a greater
 88 importance in the Amazon Basin than in other continental
 89 regimes [Roberts *et al.*, 2001; Artaxo *et al.*, 1990]. The
 90 knowledge of the vertical aerosol structures, and thus the
 91 information whether and in which way aerosols may alter
 92 cloud processes (liquid drop and ice particle formation) is
 93 essential to estimate the aerosol effects on climate. However,
 94 advanced aerosol measurements in the tropical Amazon rain
 95 forest are demanding and are constrained by the lack of
 96 infrastructure in this large inaccessible area. Long-term
 97 observations of the aerosol conditions in Amazonia have
 98 been performed by means of AERONET Sun photometer
 99 measurements [Holben *et al.*, 1996; Schafer *et al.*, 2008].
 100 But to our knowledge no continuous measurements of ver-
 101 tical aerosol profiles have been performed before 2008.

102 [8] For the first time in Amazonia, continuous aerosol
 103 observations with Raman lidar were carried out in the
 104 framework of EUCAARI (European Integrated Project on
 105 Aerosol, Cloud, Climate, Air Quality Interactions) [Kulmala
 106 *et al.*, 2011] and AMAZE-08 (Amazonian Aerosol Charac-
 107 terization Experiment) [Martin *et al.*, 2010b]. Raman lidar is
 108 of unique advantage due to two reasons. Direct extinction
 109 profiling is performed at ambient humidity conditions (i.e.,
 110 in the natural environment of aerosol layers) which is of
 111 fundamental importance for climate-impact studies. Second,
 112 the aerosol is remotely sensed and thus not manipulated
 113 before the measurement of optical and microphysical prop-
 114 erties. In the case of surface-based or airborne in situ aerosol
 115 characterization the particles are dried and the full size dis-
 116 tribution is not measured because of inlet and associated size
 117 cutoff effects. We have used the multiwavelength aerosol
 118 Raman lidar technique since 1997 and have performed sev-
 119 eral aerosol studies in polluted tropical areas in South Asia

[Franke *et al.*, 2003] and West Africa [Tesche *et al.*, 2011].
 120 In the framework of EUCAARI, Raman lidars were
 121 deployed in northern China close to Beijing [Hänel *et al.*,
 122 2012], India [Komppula *et al.*, 2012], and South Africa.
 123

[9] The paper is structured as follows: In section 2 the
 124 field site, the lidar instrument and the lidar data analysis
 125 methods are briefly described. Section 3 presents three case
 126 studies of particle optical properties, for aged smoke, young
 127 smoke, and pristine aerosol conditions observed during the
 128 wet season. Statistical results are given in section 4 in terms
 129 of layer geometrical properties describing the vertical extent
 130 of the aerosol layers and optical properties of aerosols. Sta-
 131 tistical results for the wet and the dry season are contrasted.
 132 Concluding remarks and essential findings are summarized
 133 in section 5.
 134

2. Experiment 135

2.1. Field Site 136

[10] The lidar observations were performed 60 km north
 137 of Manaus, which is on the Amazon river, at the Silvicultura
 138 research site of the National Institute for Amazonia Research
 139 (INPA). The field site at 2° 35.9'S, 60° 2.3'W and 83 m
 140 height above sea level (asl) in the central northern part of the
 141 Amazon Basin is indicated in Figure 1. The lidar was
 142 deployed on a glade surrounded by tropical rain forest which
 143 covers the sparsely populated area upwind of the lidar site for
 144 almost 1000 km. Northeasterly (wet season) to easterly and
 145 southeasterly winds (dry season) prevail throughout the
 146 entire year as a result of the trade-wind circulation. The wet
 147 season lasts from December to June and the dry season from
 148 July to November. During the dry season, a high fire activity
 149 occurs in Amazonia each year. Due to the strong easterly
 150 trade winds, advection of pollution from the two-million-
 151 inhabitants city Manaus (the so-called Manaus plume) to the
 152 field site can generally be ignored [cf. also Kuhn *et al.*,
 153 2010]. An AERONET (Aerosol Robotic Network) [Holben
 154 *et al.*, 2001] Sun photometer was mounted on a 15 m high
 155 tower at the lidar site. About 17 km west of the lidar loca-
 156 tion, the EUCAARI in situ measurements were performed
 157 using several research towers. Regular radiosondes are
 158 launched at the military airport of Manaus in the south of the
 159 city at 0000 UTC (2000 local time, LT) and 1200 UTC
 160 (0800 LT). In parallel to EUCAARI, the Amazonian Aerosol
 161 Characterization Experiment (AMAZE-08) [Martin *et al.*,
 162 2010b] took place at the same sites in February and March
 163 2008. Additional instruments for aerosol and gas investiga-
 164 tions were operated during that time.
 165

2.2. Lidar Instrument and Data Analysis 166

[11] The automated multiwavelength polarization Raman
 167 lidar Polly^{XT} (Portable Lidar System, XT indicates extended
 168 version) [Althausen *et al.*, 2009] was used for aerosol
 169 profiling. Observations were conducted from 22 January to
 170 11 November 2008 and thus covered most of the wet and dry
 171 season. During the almost 10-months observational period,
 172 lidar observations could be performed on 211 days resulting
 173 in more than 2500 hours of tropospheric aerosol and cloud
 174 profile observations. Thus, a very high data coverage in
 175 terms of observation days could be achieved. A severe laser
 176 malfunction, however, interrupted the measurements in
 177 June/July for six weeks. For the rest of the observational
 178



Figure 1. (left) Map of northern South America with lidar site (yellow star). The black line indicates the border of the rain forest. Source: <http://earthobservatory.nasa.gov>. (right) Satellite image of the experimental area showing the lidar site 60 km north of Manaus. Main wind direction from east is also indicated. Image source: Google Earth.

179 period, only minor interruptions due to, e.g., air-condition-
 180 ing problems or power failures affected our observations.
 181 [12] The advanced lidar permits us to measure vertical
 182 profiles of the particle backscatter coefficient at 355, 532,
 183 and 1064 nm, of the particle extinction coefficient at 355 and
 184 532 nm, and of the particle depolarization ratio at 355 nm.
 185 From these extensive particle properties, intensive particle
 186 properties like the extinction-to-backscatter ratio (lidar ratio)
 187 and of the Ångström exponents can be calculated. The
 188 Ångström exponent expresses the wavelength dependence
 189 of the backscatter or extinction coefficient. Microphysical
 190 properties such as the effective radius, surface-area and vol-
 191 ume concentrations, and the complex refractive index can be
 192 retrieved by means of an inversion scheme [Müller *et al.*,
 193 1999a, 1999b; Ansmann and Müller, 2005]. The obtained
 194 volume size distribution and the complex refractive index
 195 allow us to estimate the single-scattering albedo (SSA)
 196 [Müller *et al.*, 2000]. Table 1 gives an overview of the aerosol
 197 products that can be derived from Polly^{XT} measurements.
 198 [13] For the independent determination of particle back-
 199 scatter and extinction coefficients, the Raman lidar method
 200 [Ansmann *et al.*, 1992] is applied. At daytime, when the
 201 Raman channels cannot be used, particle backscatter coef-
 202 ficients are retrieved with the Fernald algorithm [Fernald,
 203 1984]. The volume and particle linear depolarization ratio
 204 are determined by following the method of Murayama *et al.*
 205 [1999]. Lidar signals are influenced by particle and Rayleigh
 206 backscatter and light-extinction processes. For the removal
 207 of Rayleigh scattering effects, molecular backscatter and
 208 extinction coefficients are computed after Bucholtz [1995]
 209 by using temperature and pressure profiles as measured
 210 with radiosonde launched at the Manaus military airport
 211 twice a day.
 212 [14] More than 50 nighttime observations were available
 213 from the dry season measurements for extended aerosol

profile studies using the Raman lidar technique. During the
 214 wet season, the prevalence of clouds, rain and fog made
 215 observing difficult. As a consequence, only the retrieval of
 216 the backscatter coefficients and the depolarization ratio was
 217 possible for most of the wet season measurements cases.
 218 [15] Large uncertainties in the laser-beam receiver-field-
 219 of-view overlap correction in the lowest several hundreds of
 220 meters restrict the retrieval of the particle backscatter pro-
 221 files to heights above about 400 m (Raman lidar method) or
 222 750 m above ground level (agl) (Fernald method). Reliable
 223 particle extinction coefficients obtained with the Raman
 224 lidar method are available for heights above 1000 m after
 225 overlap correction by means of the method of Wandinger
 226 and Ansmann [2002]. The full overlap between the field of
 227 view of the telescope and the laser beam is achieved at
 228 1500 m height. In summary, retrieval uncertainties are of the
 229 order of 5%–10% (backscatter coefficient, depolarization
 230

Table 1. Overview of the Lidar-Derived Aerosol Products^a t1.1

Parameter	Symbol	Remarks	t1.3
Backscatter coefficient	β	355, 532, 1064 nm	t1.4
Extinction coefficient	α	355, 532 nm	t1.5
Linear depolarization ratio	δ	355 nm	t1.6
Lidar ratio	$S = \frac{\alpha}{\beta}$	355, 532 nm	t1.7
Ångström exponent	\hat{a}	for α and β	t1.8
Volume size distribution		via inversion	t1.9
Effective radius	r_{eff}	via inversion	t1.10
Number, surface, and volume concentration		via inversion	t1.11
Complex refractive index		via inversion	t1.12
Single-scattering albedo		inversion + Mie code	t1.14

^aAn inversion scheme is used to derive microphysical properties and
 complex refractive index from the measured particle optical properties.
 Single-scattering albedo can be computed by means of a Mie scattering
 algorithm from the inversion products.

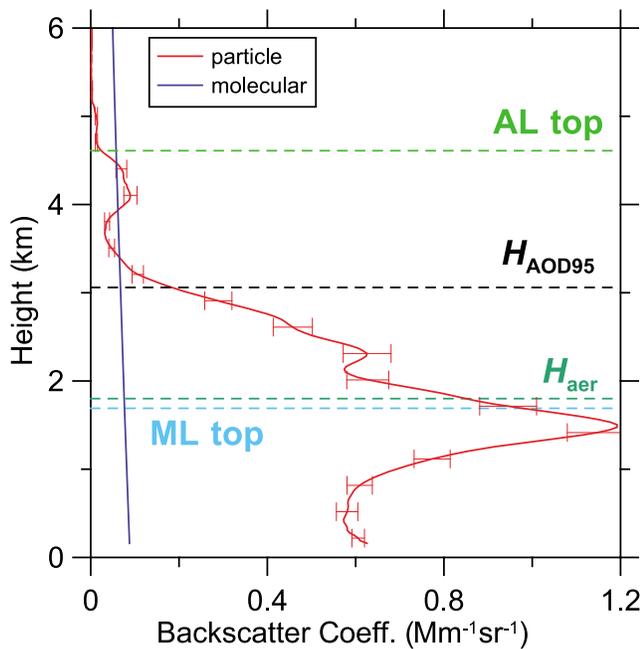


Figure 2. Four different layer height parameter used to characterize aerosol layering over the Amazon Basin. Aerosol layer height AL top, optical-depth-related scale height H_{aer} , H_{AOD95} (height at which 95% of the total AOD is reached), and ML top (top of the mixing layer as derived from ECMWF) are indicated as horizontal dashed lines. The vertical profile of the 1064 nm backscatter coefficient observed on 08 September 2008 is shown as measurement example (red curve with error bars).

231 ratio), 10%–20% (extinction coefficient), and 20%–30%
232 (lidar ratio).

233 [16] The retrieval of the particle optical depth (AOD) was
234 performed in the following way. When the profile of the
235 particle extinction coefficient was available (dry season),
236 these data were used in the AOD computation for the height
237 range >1000 m. For heights <1000 m, the profile of the
238 backscatter coefficient was taken. The particle backscatter
239 coefficients were multiplied with the most appropriate lidar
240 ratio, which is given by the Raman lidar observations of
241 extinction and backscatter coefficient at heights in the upper
242 part of the mixed layer (1–1.5 km height). For the lowermost
243 400 m for which no reliable backscatter coefficients are
244 available, we assume height-independent backscattering and
245 use the backscatter values for 400 m throughout the layer
246 from the surface to 400 m height. This procedure leads to
247 uncertainties in the AOD values of the order of 10% (during
248 the dry season) as our sensitivity studies and comparisons
249 with Sun photometer observations show. For the wet season
250 measurements, the AOD retrieval is completely based on the
251 backscatter coefficient profiles. A lidar ratio of 60 sr was
252 generally assumed in the estimation of the particle extinction
253 profile from the backscatter profile. The AOD uncertainty is
254 then of the order of 25%–30% according to realistic varia-
255 tions in the lidar ratio from 40 to 80 sr.

256 [17] From the vertical profiles of the backscatter and
257 extinction coefficients several geometrical parameter were
258 derived, as illustrated in Figure 2. AL top is defined as the

height, at which the 1064 nm particle backscatter coefficient 259
drops below the threshold value of $0.02 \text{ Mm}^{-1} \text{ sr}^{-1}$ for the 260
first time as a function of height. This threshold backscatter 261
coefficient corresponds to a 1064 nm extinction coefficient 262
of about 1 Mm^{-1} and a particle-to-Rayleigh extinction ratio 263
of about 2 at 4 km height. H_{AOD95} is defined as that height at 264
which 95% of the total AOD are caused by particles in the 265
tropospheric layer below H_{AOD95} . The AOD scale height 266
 H_{aer} is defined as the height at which about 37% of the AOD 267
($1/e$ AOD) is caused by particles above H_{aer} . The mixing- 268
layer top (ML top) is derived from European Centre for 269
Medium-Range Weather Forecasts (ECMWF) model runs. 270
These ML-top data are kindly provided by the Finnish 271
Meteorological Institute. 272

[18] As a final remark, several efforts were undertaken to 273
meet the hardware and software standards of the European 274
Research Lidar Network (EARLINET) [Pappalardo *et al.*, 275
2010]. Calibration procedures were performed at the field 276
site as well as afterwards at Leipzig, Germany, and applied, 277
e.g., in the corrections of overlap effects and polarization 278
effects in the receiver unit. These efforts as well as com- 279
parisons to other lidars and instruments (i.e., Sun photome- 280
ter) showed that the measured aerosol profiles obtained with 281
Polly^{XT} are of high quality and fulfill EARLINET standards 282
[Pappalardo *et al.*, 2010, and references therein]. 283

3. Case Studies 284

[19] Two cases with fresh (local) and aged smoke 285
(regional haze) observed during the dry season are discussed 286
in detail. They show very different optical and microphysical 287
properties and thus provide an impression of the broad 288
spectrum of smoke characteristics. As a contrast to the bio- 289
mass burning cases, an observation at pristine conditions 290
during the wet season with an AOD of less than 0.02 is 291
presented in addition. 292

3.1. Aged Smoke 293

[20] Reid *et al.* [1998] provides a detailed explanation of 294
the formation of aged biomass burning smoke in the Ama- 295
zon Basin. After being emitted, the smoke particles disperse 296
and have the potential to be rapidly transported into the 297
lower atmosphere up to the strong trade wind inversion at a 298
height of about 3–4 km as a result of the high air tempera- 299
tures during emission. Smoke from hundreds of fires mix 300
with biogenic emissions from forests and suspended soil 301
particles (and potentially with urban haze). During transport, 302
smoke undergoes photochemical transformations, gas-to- 303
particle conversion, and particle coagulation. Smoke can be 304
entrained into clouds where increased efficiencies of specific 305
chemical reactions may accelerate the growth of the smoke 306
particles. 307

[21] Based on their observations during SCAR-B (Smoke, 308
Clouds and Radiation - Brazil), Reid *et al.* [1998] found that 309
condensation and gas-to-particle conversion of inorganic 310
and organic vapors increase the aerosol mass by 20%–45%. 311
30%–50% of this mass growth likely occurs in the first few 312
hours. The remaining mass growth is probably associated 313
with photochemical and cloud-processing mechanisms 314
operating over several days. After three days, most of the 315
condensation and gas-to-particle conversion has likely been 316
taken place [Reid *et al.*, 1998]. Coagulation is then left to be 317

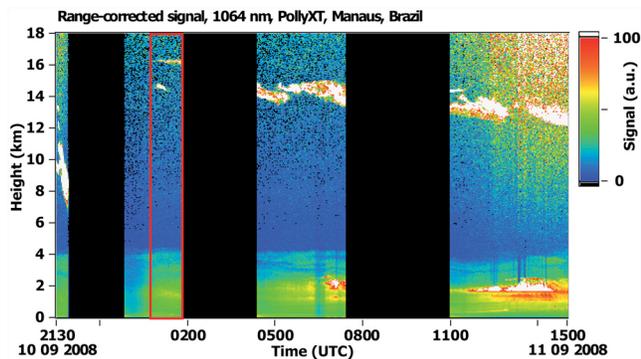


Figure 3. Aerosol layering observed from 10 September 2008, 2130 UTC (1930 LT) to 11 September 2008, 1500 UTC (1100 LT). The range-corrected signal at 1064 nm wavelength is shown. White features indicate low level clouds (around 2 km height agl) and ice clouds (mostly between 12 km height and the tropopause above 16 km height). The red box indicates the signal averaging period for an in-depth study of particle optical and microphysical properties.

318 the only significant particle growth mechanism over the next
319 days of long-range transport.

320 [22] Müller *et al.* [2007a] investigated the growth of bio-
321 mass burning particles as a function of travel time. They
322 observed that the surface-area-weighted radius (effective
323 radius) increases from values of 0.1 μm (1 day after emis-
324 sion), 0.15–0.25 μm (2–4 days after the emission) to values
325 of 0.3–0.4 μm after 10–20 days of travel time. As shown by
326 lidar observations [Mattis *et al.*, 2003] and subsequent
327 model studies [Damoah *et al.*, 2004], fire smoke can survive
328 over weeks in the free troposphere before it is removed by
329 washout processes.

330 [23] The composition of the aerosol in the Amazon Basin
331 can be divided into five possible components according to

Reid et al. [1998]: primary smoke products, secondary smoke
332 products, other anthropogenic materials, biogenic materials,
333 and soils. The relative contribution of these components to
334 the aerosol mass loading is highly variable. Continuously
335 occurring changes in the particle size distributions and
336 compositions during aging have a large impact on the optical
337 properties of the aerosol. Different burning types (flaming
338 and smoldering fires) increase the complexity of observed
339 microphysical, chemical, and optical properties of smoke
340 plumes.
341

[24] Our first case study deals with aged smoke. The lidar
342 observations on 10 and 11 September 2008 are shown in
343 Figure 3. A dense aerosol layer extended up to about 4 km.
344 Cirrus occurred in addition, mainly in the upper troposphere.
345 Fog (in the lowermost few hundred meters) occurred and
346 prohibited high quality lidar observations from 0000–
347 0030 UTC (2000–2030 local time, LT). Fog formation also
348 attenuated the laser beam significantly between 0630 and
349 0700 UTC. The evolution of low clouds around 2 km height
350 started after 0700 UTC on 11 September 2008.
351

[25] According to AERONET Sun photometer observations
352 in the late afternoon on 10 September 2008 (2039 UTC), the
353 total and fine-mode 500 nm AOD were 0.45 and 0.43 (fine-
354 mode fraction of 95%). The 500 nm AOD increased to 0.6 in
355 the morning of 11 September 2008 (1223 UTC). Photometer-
356 derived Ångström exponents around 1.2 and effective radii of
357 0.23–0.26 μm on the late afternoon on 10 September 2008 are
358 indicative for aged smoke [Reid *et al.*, 1998].
359

[26] Figure 4 (right) shows the 550 nm AOD over the
360 Amazon Basin retrieved from MODIS (Moderate Resolution
361 Imaging Spectroradiometer) [Remer *et al.*, 2005] observa-
362 tions on 10–11 September 2008. According to the satellite
363 measurements, an aerosol plume was located southeast of
364 the lidar site. AOD values up to 0.74 in southern Amazonia
365 and of 0.5 for the Manaus region, respectively, were found.
366

[27] A very pronounced fire activity in the south, south-
367 east, and east of the lidar site obviously caused this aerosol
368 plume over Amazonia (see Figure 4, left). Fire counts for
369

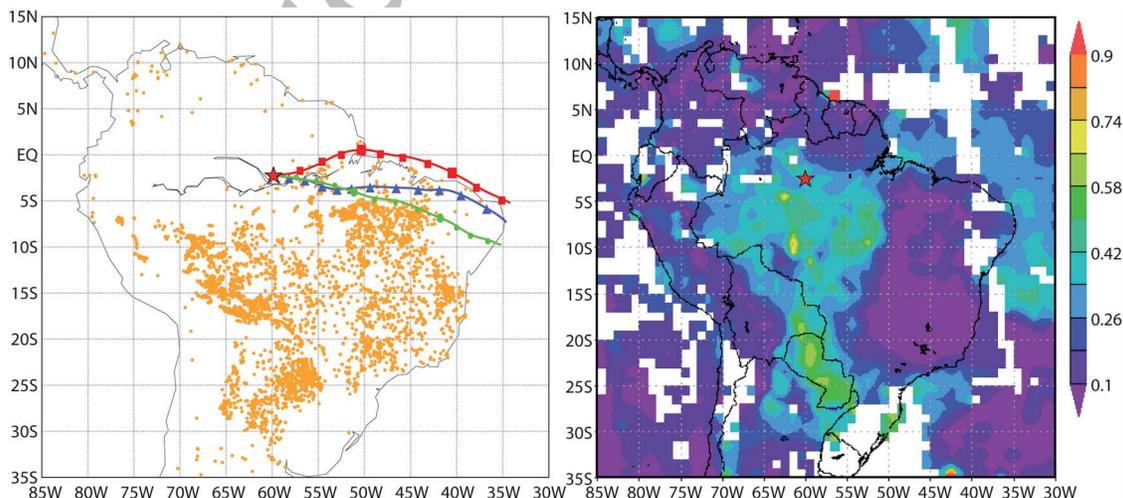


Figure 4. (left) Fire counts (orange dots) as derived by FIRMS for 7–10 September 2008 and HYSPLIT backward trajectories in 6-h steps (indicated by symbols) for the arrival heights of 1500 (blue), 3500 (red), and 5000 m agl (green) for 11 September 2008, 0100 UTC. (right) MODIS AQUA AOD composite (550 nm) for 10–11 September. The red star indicates the lidar site.

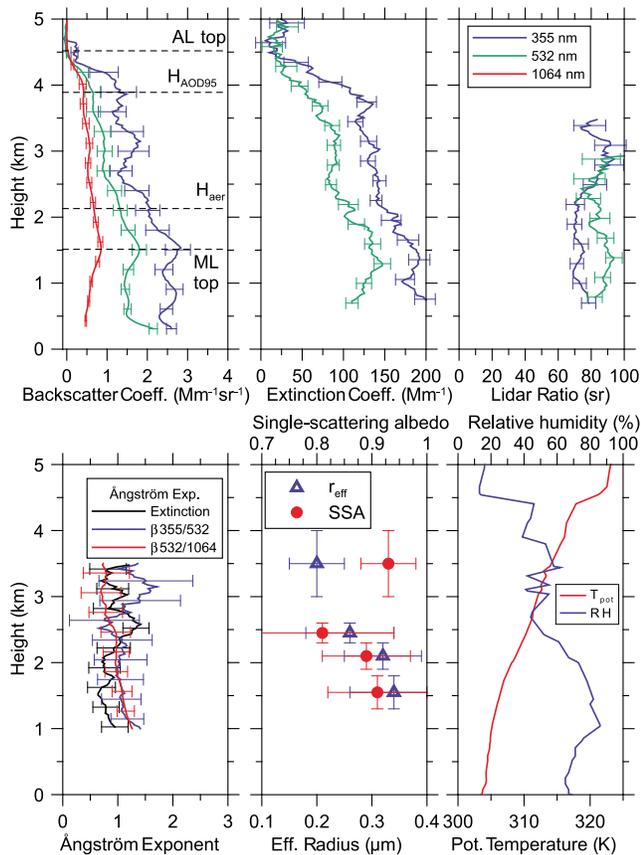


Figure 5. Vertical profiles of particle backscatter coefficient, extinction coefficient, and lidar ratio for several wavelengths, Ångström exponents, effective radius (r_{eff}), and single-scattering albedo (SSA) observed on 11 September 2008, 0100 UTC–0200 UTC (2100–2200 LT). The 532 nm AOD is 0.44. Potential temperature (T_{pot}) and relative-humidity (RH) profiles were measured with Manaus radiosonde launched on 11 September 2008, 0000 UTC. Different aerosol layer heights (AL top, H_{AOD95} , H_{aer} , ML top) are indicated by horizontal lines in the backscatter panel. Before the computation of the optical properties, lidar signals are vertically smoothed with window lengths of 270 m (backscatter), 750 m (extinction), and 990 m (lidar ratio). Layer mean values of effective radius and SSA are determined by inversion. Vertical bars indicate the layer depth.

370 7–10 September 2008 as obtained from MODIS measure-
 371 ments (via Fire Information for Resource Management Sys-
 372 tem (FIRMS) at University of Maryland [Giglio *et al.*, 2003])
 373 and 3-day backward trajectories for the arrival heights of
 374 1500, 3500, and 5000 m from HYSPLIT (Hybrid Single
 375 Particle Lagrangian Integrated Trajectory Model, [http://](http://ready.arl.noaa.gov/HYSPLIT.php)
 376 ready.arl.noaa.gov/HYSPLIT.php) are shown in Figure 4
 377 [Draxler and Hess, 1998; Draxler *et al.*, 2009]. Meteorolo-
 378 gical fields from the archived model assimilation data sets
 379 of GDAS (NCEP Global Data Assimilation System) were
 380 used. The backward trajectories indicate an air-mass flow
 381 from easterly directions. The air masses crossed fire-active
 382 regions 1–2 days before the arrival at the lidar site. Freshly

emitted smoke was added to the obviously already aged 383
 biomass burning aerosol (regional haze). 384

[28] The vertical profiles of the optical and microphysical 385
 properties of the smoke aerosol as observed with our lidar 386
 are presented in Figure 5 for the observation period between 387
 0100 and 0200 UTC (indicated by a red box in Figure 3). 388
 Particle backscatter and extinction coefficients for the trans- 389
 mitted laser wavelengths, respective particle lidar ratios, 390
 Ångström exponents, effective radii, and SSA values (532 nm) 391
 determined for this one-hour period are shown. The vertical 392
 profiles of relative humidity and potential temperature 393
 derived from observations with radiosonde launched at the 394
 Manaus military airport at 11 September 2008, 0000 UTC, 395
 are given in addition. 396

[29] AL top height, H_{AOD95} , and the optical-depth-related 397
 scale height H_{aer} are 4.5, 3.9, and 2.1 km, respectively. The 398
 maximum mixing layer height ML top was about 1.6 km 399
 on 10 September 2008. AL top coincides with a strong tem- 400
 perature inversion (trade wind inversion layer). A moist 401
 atmosphere with relative humidities of 60%–80% within the 402
 lowermost 2.5 km of the troposphere, and 40%–50% from 403
 2.5–4.5 km height was observed. 532 nm particle extinction 404
 coefficients were 150–200 Mm^{-1} for heights <2.5 km in the 405
 moist air, and 100–150 Mm^{-1} in the drier air. The similarity 406
 of the relative humidity and the extinction profiles indicate 407
 water uptake by the particles. The extinction coefficient for 408
 Amazonian smoke roughly increases by a factor of 1.5 when 409
 the relative humidity increases from 40% to 80% [Rissler 410
et al., 2006]. All in all, an almost vertically homogeneous 411
 haze layer was observed up to 4 km height. Ångström 412
 exponents are of the order of 1 and are thus in agreement 413
 with the Sun photometer observations. 414

[30] It is worth to mention that the observed particle 415
 depolarization ratio [Baars *et al.*, 2011] was always very low 416
 (<0.03) throughout the dry season. This observation corro- 417
 borates the assumption that aged water-containing biomass 418
 burning particles can be regarded to be spherical. 419

[31] The lidar ratios of 70–90 sr shown in Figure 5 are 420
 indicative for considerably light-absorbing smoke particles 421
 [Franke *et al.*, 2003; Müller *et al.*, 2007b; Tesche *et al.*, 422
 2011]. The larger lidar ratios at 532 nm compared to the 423
 ones for 355 nm in the main haze layer (below 2.5 km 424
 height) are another indication for the aged smoke (4–10 days 425
 old biomass burning particles) [Müller *et al.*, 2005; Ansmann 426
et al., 2009]. 427

[32] The particle effective radius of about 0.3 μm in the 428
 main and humid aerosol layer below 2.5 km is roughly a 429
 factor of 2 larger than respective values found by Reid *et al.* 430
 [1998] for dry Amazonian smoke particles. Water uptake 431
 effects are responsible for these large effective radii on 432
 11 September 2008. The effective radius decreases with 433
 height and is 0.2 μm in the drier air in upper part of the 434
 smoke layer. 435

[33] The lidar-derived SSA of the water-containing smoke 436
 particles of around 0.9 in the main layer below 2.5 km height 437
 are consistent with the values found by Reid *et al.* [1998] for 438
 dry smoke particles (0.8–0.9 for 550 nm) in southeastern 439
 Amazonia. The mass fraction of black carbon in the Ama- 440
 zonian smoke aerosol is in the range of 5%–10% [Reid *et al.*, 441
 1998, 2005], but may vary from 2%–30% [Reid and Hobbs, 442
 1998; Reid *et al.*, 2005]. Tesche *et al.* [2011] presented SSAs 443
 of, on average, even <0.8 (at 532 nm) for highly absorbing 444

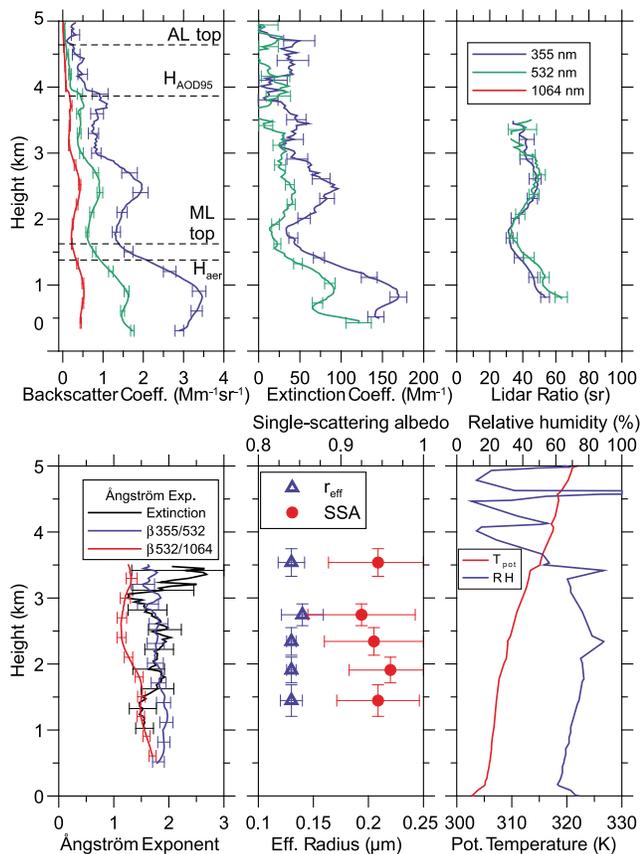


Figure 6. Same as Figure 5 except for 15 August 2008, 2235–2335 UTC (1835–1935 LT). AOD (532 nm) is 0.15. Potential temperature (T_{pot}) and relative humidity (RH) profiles were measured with Manaus radiosonde launched on 16 August 2008, 0000 UTC.

445 African smoke (at relative humidities below 60% in the
446 smoke layers). *Dubovik et al.* [2002] reported AERONET-
447 photometer-derived values from 0.9–0.94 for Amazon for-
448 est-fire smoke and 0.86–0.92 for South American savanna
449 smoke (at ambient humidity conditions).

450 [34] In the drier layer between 3 and 4 km in Figure 5, the
451 SSA slightly increased to values around 0.93. Different
452 burning characteristics (smoldering versus flaming fires),
453 differences in the burning material and thus composition of
454 the smoke particles, and transport time may be responsible
455 for the differences in the smoke optical properties, effective
456 radius, and single-scattering albedo observed with the lidar
457 above and below 2.5 km height.

458 [35] Finally, we estimated the mass-specific extinction
459 coefficients and smoke mass concentrations based on com-
460 bined photometer-lidar observations [*Ansmann et al.*, 2012].
461 In the retrieval, a density of the smoke particles of 1.35 g/cm^3
462 is assumed [*Reid and Hobbs*, 1998; *Reid et al.*, 2005].
463 The specific extinction coefficients are around $4 \text{ m}^2/\text{g}$
464 (AERONET photometer, evening of 10 September 2008)
465 and $3.5 \text{ m}^2/\text{g}$ (lidar, moist layer below 2.5 km height) and
466 $4.5 \text{ m}^2/\text{g}$ (lidar, dry layer above 2.5 km height). With
467 increasing water content (and thus decreasing particle density
468 toward 1 g/cm^3) the mass-specific extinction coefficient
469 increase (e.g., toward 6 instead of $4.5 \text{ m}^2/\text{g}$). Reid derived

mass-specific extinction coefficients of $4 \pm 1 \text{ m}^2/\text{g}$ (at
470 550 nm) for dry Amazonian smoke particles. By using a
471 specific extinction coefficient of $4 \text{ m}^2/\text{g}$ we obtain particle
472 mass concentrations of $30\text{--}40 \mu\text{g/m}^3$ in the main part of the
473 smoke haze layer below 2.5 km height, and a value around
474 $15 \mu\text{g/m}^3$ for the dry layer from 3–4 km height. 475

3.2. Young Smoke 476

[36] A case dominated by freshly emitted smoke was
477 observed in early evening of 15 August 2008 (1835–
478 1935 LT). Lidar profiles of optical and microphysical
479 properties are shown in Figure 6. *Reid et al.* [1998] men-
480 tioned that smoke emissions in Brazil have a strong diurnal
481 cycle. Fires are generally ignited in the late morning through
482 late afternoon. Thus haze sampled in the early evening are
483 most likely to contain a large fraction of young smoke. 484

[37] The optical properties show a distinct layering of
485 particles. AL top was close to 4.5 km and the optical-depth
486 scale height H_{aer} at about 4.5 km and the optical-depth
487 maximum ML top in the afternoon of 1600 m. 532 nm
488 particle extinction coefficients ranged from $20\text{--}120 \text{ Mm}^{-1}$
489 in the lowermost 3 km of the atmosphere. The 532 nm
490 optical depth was 0.15. AERONET photometer observations
491 are not available for this case because of persistent cirrus
492 layers. 493

[38] The Ångström exponents were significantly higher
494 than on 11 September 2008 (aged smoke case) with values
495 of 1.5–2 for the wavelength range from 355–532 nm. Cor-
496 respondingly, the effective radius was small with values
497 around $0.13 \mu\text{m}$. If we take the water-uptake effect into
498 account (relative humidities ranged from 60–90% in the
499 lowermost 3.5 km), the dry particle effective radius was
500 certainly clearly below $0.1 \mu\text{m}$. According to *Reid et al.*
501 [1998], the high Ångström exponents of 1.5–2 and the
502 very low effective radii point to freshly emitted smoke. 503

[39] The lidar ratios showed surprisingly low values for
504 fresh smoke. We again expected highly absorbing particles
505 and thus values $>70 \text{ sr}$. The lidar ratio increases not only
506 with increasing particle absorption but also with decreasing
507 particle size. Such low values of $30\text{--}60 \text{ sr}$ together with the
508 high Ångström exponents indicate weakly absorbing parti-
509 cles. The negligible wavelength dependence of the lidar ratio
510 is another characteristic for fresh smoke. The reason for
511 these unusually low lidar ratios remains unclear. However,
512 according to *Reid and Hobbs* [1998] and *Reid et al.* [2005],
513 the black carbon content can vary from 2%–30%. *Müller*
514 *et al.* [2005] presented statistics for Canadian and Siberian
515 forest fire smoke (after travel times of >6 days) and also
516 found lidar ratios spanning a large range from 30 to 90 sr. In
517 agreement with the rather low lidar ratios, the SSA is high
518 with values of 0.92–0.95 (see Figure 6, bottom). 519

[40] *O'Neill et al.* [2002] presents a Sun photometer study
520 of aerosol properties at ambient conditions of boreal forest
521 fires in western Canada. Several photometers were close to
522 the fire sources (30–600 km), and others far way ($>2000 \text{ km}$).
523 For the small distances, the smoke-related fine-mode
524 Ångström, effective radii, and single-scattering albedo, were
525 1.5–2.5, $0.13\text{--}0.17 \mu\text{m}$, and mostly >0.95 , respectively. For
526 the large distances (aged smoke), they found lower Ångström
527 exponents (1–1.5) and lower single scattering albedos
528 (mostly <0.95). The effective radii were similar for the
529 both data sets. These findings for boreal forest fires are 530

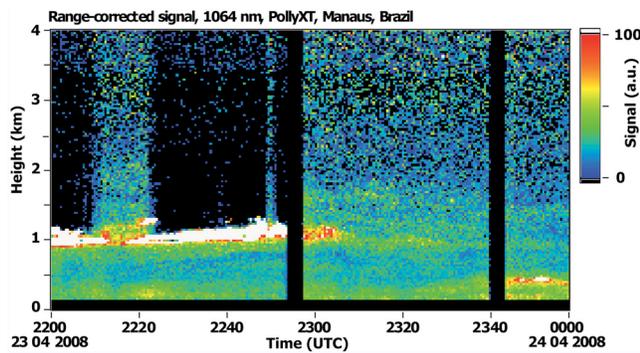


Figure 7. Temporal evolution of aerosol layering on 23 April 2008, 2200–2400 UTC (1800–2000 LT), in terms of the range-corrected 1064 nm signal. The mean 532 nm AOD for the 2300–0000 UTC period was estimated to be 0.019.

531 qualitatively in good agreement with our lidar observations
532 of aged and young smoke.

533 3.3. Pristine Conditions in the Wet Season

534 [41] The main results of the lidar observations performed
535 during the wet season are discussed by *Baars et al.* [2011].
536 For the first time, clear and unambiguous indications for a
537 significant long-range transport of African biomass burning
538 aerosol to the Amazon Basin were documented [*Ansmann*
539 *et al.*, 2009; *Baars et al.*, 2011].

540 [42] Here, we present one case obtained for rather pristine
541 conditions to emphasize the strong contrast between natural
542 aerosol conditions and man-made haze and smog situations
543 in the Amazon Basin. Figure 7 shows the aerosol layering
544 measured with lidar on 23 and 24 April 2008. Rain associ-
545 ated with strong washout effects was not observed during the
546 complete lidar measurement session starting at 1800 UTC
547 (1400 LT). In the beginning of the analyzed time period (at
548 2200 UTC), low-level clouds were present at around 1 km
549 agl which prohibited the penetration of the laser beam to
550 higher altitudes. At about 2300 UTC, the low-level clouds
551 dissolved and the vertical extent of the aerosol layer of 2 km
552 became visible to the lidar.

553 [43] The vertical profiles of the particle backscatter coef-
554 ficient at 532 and 1064 nm and the respective Ångström
555 exponent for the cloud-free period after 2330 UTC are shown
556 in Figure 8. One aerosol layer near to the surface and a sec-
557 ond aerosol layer centered at around 1 km height were
558 observed. In the higher layer the low level clouds occurred.
559 The particle backscatter coefficients multiplied with a lidar
560 ratio of 60 ± 20 sr provide estimates for the 532 nm extinc-
561 tion coefficients. Values of $5\text{--}15 \text{ Mm}^{-1}$ are rather low and
562 indicate pristine conditions. The 532 nm AOD of $0.019 \pm$
563 0.008 was estimated from the extinction profile. The Ång-
564 ström exponents of around 1.5 are typical for accumulation
565 mode particles. Because of instrumental problems, no infor-
566 mation from the UV channels was available on that day.

567 [44] The AOD of 0.019 is lower than the average AOD
568 over clean marine sites [*Smirnov et al.*, 2009], and is one of
569 the lowest values ever measured over a continental site.
570 Measurements near Antarctica showed similar low AOD
571 values [*Holben et al.*, 2001; *Wilson and Forgan*, 2002].

[45] The maximum ML top on that day was calculated to 572
be 760 m. Thus, it was slightly higher than the observed 573
top of the first aerosol layer. The AOD scale height H_{aer} 574
was 960 m. The AL top and the height H_{AOD95} coincide at 575
1750 m. 576

[46] The low observed AOD value is even more remark- 577
able when taking the high relative humidity of $>80\%$ – 90% 578
into account. Natural Amazonian aerosol was classified as 579
moderately hygroscopic in previous experiments [*Zhou et al.*, 580
2002; *Rissler et al.*, 2004] so that hygroscopic growth should 581
enhance the aerosol light scattering. In summary, one can 582
conclude that the observed aerosol conditions on 23 April 583
2008 with an AOD of 0.02 represent background or natural 584
aerosol conditions over the Amazon rain forest. 585

[47] Such pristine conditions with an AOD < 0.05 at 586
532 nm were observed in about 50% out of all measurement 587
cases during the wet season. Aerosol was then trapped in the 588
lowermost 2.5 km of the troposphere. However, in about one 589
third of all measurements advection of smoke and dust 590
aerosol from Africa was observed as discussed by *Baars* 591
et al. [2011]. AODs ranged then typically from 0.07–0.25. 592

4. Statistical Results: Seasonal Aerosol Characteristics

[48] In this section, the essential results of the statistical 595
analysis of the lidar measurements in the dry and wet season 596
2008 are presented and discussed. The general wind patterns 597
prevailing in 2008 are analyzed in section 4.1. The geo- 598
metrical properties are given in section 4.2. The statistical 599
findings for the particle optical properties (backscatter and 600
extinction coefficients, lidar ratio, Ångström exponents, 601
particle optical depth) are then discussed in section 4.3. 602

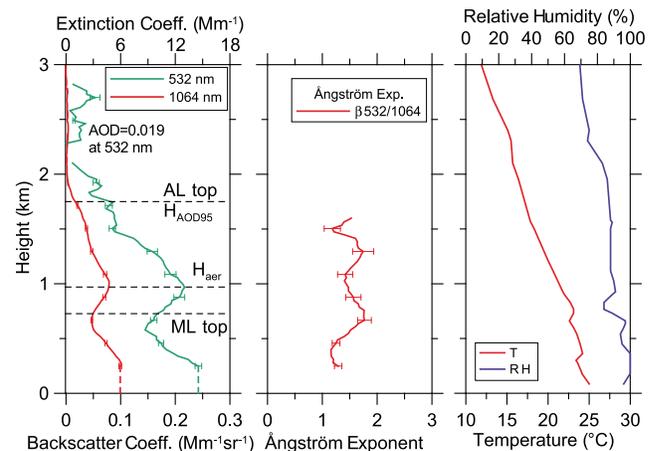


Figure 8. Vertical profiles of the 532 nm (green) and 1064 nm particle backscatter coefficient (red) and corresponding Ångström exponent measured on 23 April 2008, 2330–0000 UTC (1930–2000 LT). Extinction coefficient values (upper scale in the left panel) are obtained by multiplying the backscatter coefficient with a lidar ratio of 60 sr. The 532 nm AOD is estimated from the backscatter profile to be 0.019. Temperature (T) and relative humidity (RH) profile were measured with Manaus radiosonde on 24 April 2008, 0000 UTC. Different layer heights are indicated by horizontal lines in left panel.

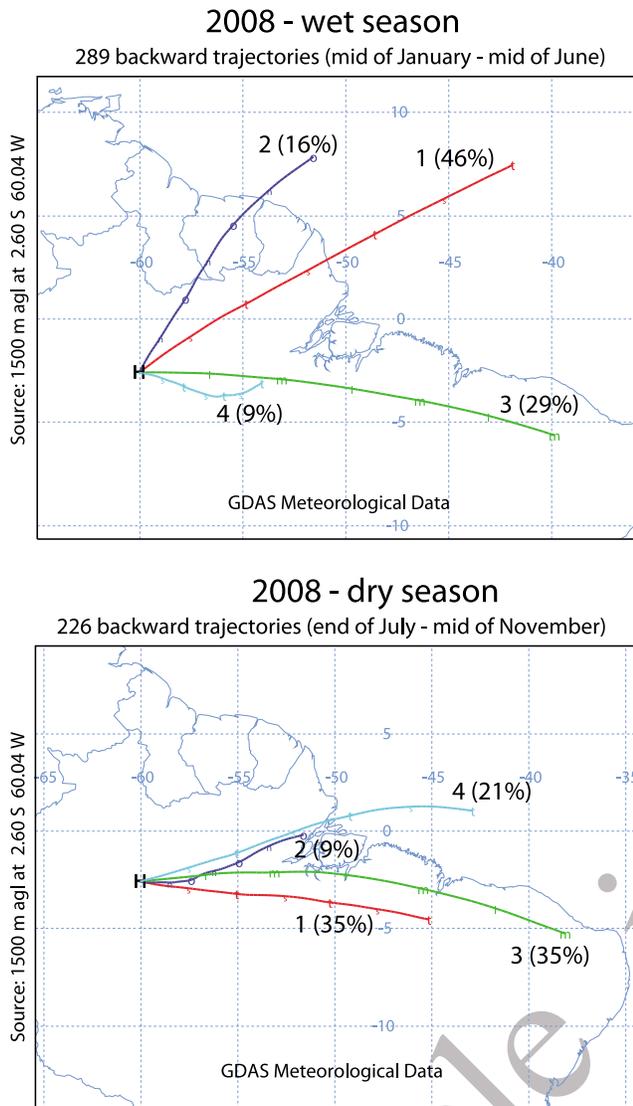


Figure 9. Cluster analysis of backward trajectories for the wet and dry season based on daily 172-h backward trajectories at an arrival height of 1500 m agl. Four clusters for each season were identified. The frequency of occurrence of each of the clusters (1–4) is given in brackets. Two trajectories per day (for arrival times of 0000 and 1200 UTC) were calculated.

603 **4.1. Meteorological Conditions**

604 [49] A cluster analysis (offline-version of HYSPLIT, 605 version 4.9, <http://ready.arl.noaa.gov/HYSPLIT.php>) based 606 on HYSPLIT backward trajectories for Manaus arrival height 607 of 1500 m agl and arrival times of 0000 and 1200 UTC for 608 each day from mid January to mid June 2008 (wet season) 609 and from end of July to mid November 2008 (main part of 610 the dry season) was performed. As shown in Figure 9, the 611 analysis revealed that air masses were transported from 612 easterly directions to the field site during both seasons as a 613 result of the prevailing trade-wind circulation. Four clusters 614 for each season were identified. In the wet season, mostly 615 north-easterly air flows occur (in 62% out of all cases),

616 whereas the air masses came mainly from easterly to south- 617 easterly direction in the dry season (in 70% out of all cases). 618 [50] The backward-trajectory analysis also suggests that 619 the impact of the Manaus pollution plume on the aerosol 620 conditions at the field site was rare. These findings corroborate 621 the results presented by *Kuhn et al.* [2010]. The authors 622 showed that the Manaus pollution plume is usually transported 623 to southwesterly directions. The dispersion of the Manaus 624 plume was found to be low so that only regions directly 625 downwind of the city were affected.

626 **4.2. Layer Geometrical Characteristics**

627 [51] Figure 10 shows histograms for the aerosol layer top 628 height (AL top) and the AOD scale height (H_{aer}) separately 629 for the wet and dry season. As can be seen, the top heights 630 accumulated between 2 and 3 km during the wet season 631 2008. A broad distribution was found for the dry season with 632 most values in the range from 3–5 km height. AOD scale 633 heights H_{aer} were not very different during the wet and dry 634 season. The main aerosol layer (below H_{aer}) reached to 500– 635 1500 m (wet season) and 1000–2000 m (dry season) most of 636 the time.

637 [52] Figure 11 shows time series of AL top, H_{aer} , and 638 H_{AOD95} in 2008. In terms of the different layer depths a 639 pronounced shift toward higher values and larger spread of 640 the layer depths during the dry season is visible. However, 641 H_{aer} shows less variability than the AL top height. Mean 642 values of AL top, H_{AOD95} , H_{aer} , and ML top are 4.1, 3.0, 1.6, 643 and 1.5 km for the dry season, and 2.5, 2.3, 1.2, and 1.1 km 644 for the wet season. The AOD scale height almost coincides 645 with mixing layer height during both seasons. The depth of 646 the layer which causes 95% of the AOD was almost the 647 same as the aerosol top height during the wet season, but 648 was often considerably below the height at which the last 649 traces of particles are detected during the dry season. On 650 average, AL top in the dry season was 1.5 km higher than in 651 the wet season whereas the main contribution to the particle 652 optical depth stems from the particle below about 1.5 km 653 disregarding the season.

654 **4.3. Particle Optical Properties**

655 **4.3.1. Backscatter Profiling**

656 [53] Figure 12 shows mean profiles of the 532 nm particle 657 backscatter coefficient for pristine conditions during the wet 658 season, for periods with advection of African dust and smoke 659 during the wet season, and for the dry season. Cloud- 660 screened observations are considered only. All individual 661 one or two-hour mean backscatter profiles observed on more 662 than 50 different days during the dry season are shown to 663 indicate the strong variability of the optical properties as a 664 function of burning intensities, atmospheric layering conditions, 665 and varying relative humidity conditions. Throughout 666 the entire dry and wet seasons, aerosol layers above 6.5 km 667 were not detected. Compared to pristine condition in the wet 668 season, the backscatter coefficients are, on average, enhanced 669 by a factor of 6. However, during events with African aerosols, 670 the particle backscatter level is also clearly larger (by a 671 factor of 4) compared to undisturbed Amazonian aerosol 672 conditions [*Baars et al.*, 2011].

673 [54] Histograms of the backscatter-related Ångström 674 exponents in the dry season (Figure 13) peak at values

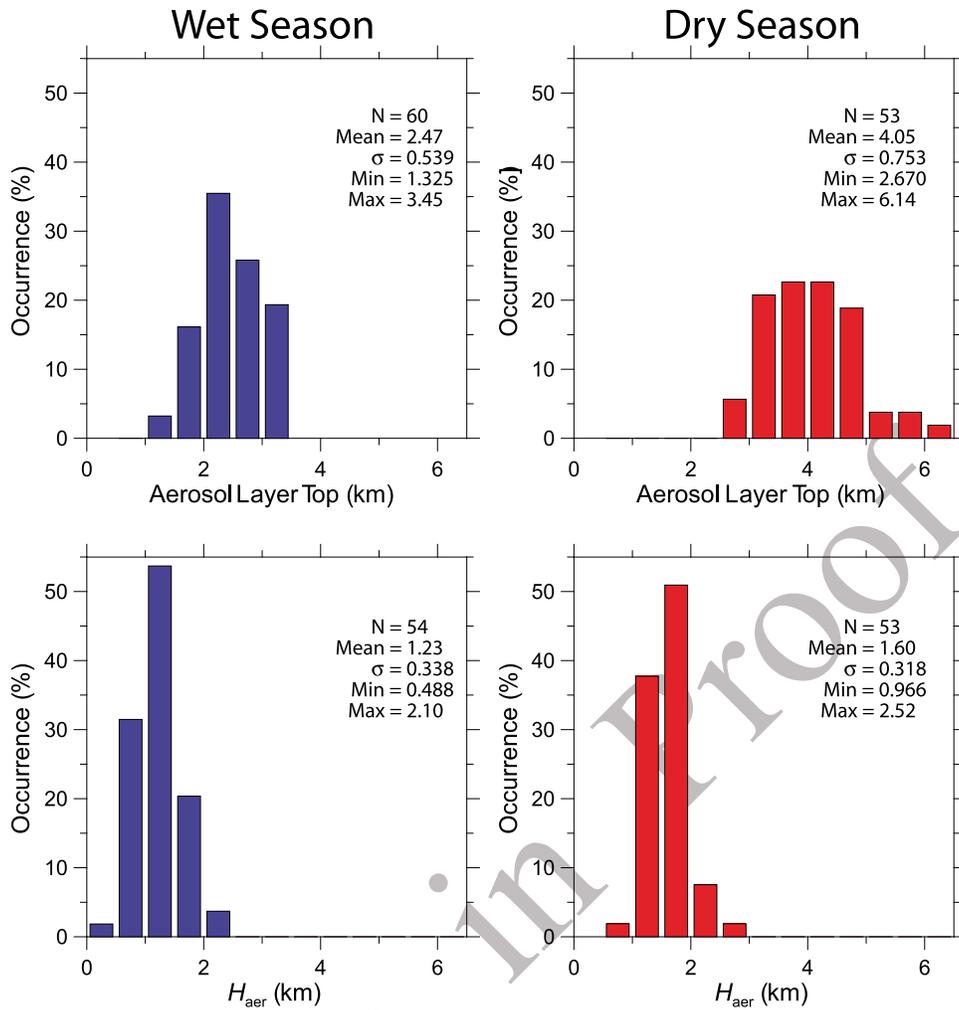


Figure 10. Frequency distribution of (top) AL top and (bottom) optical-depth-related scale height H_{aer} for the wet season (blue) and the dry season (red). The statistics are based on N observations. Mean values, standard deviations, maximum and minimum values are given as numbers.

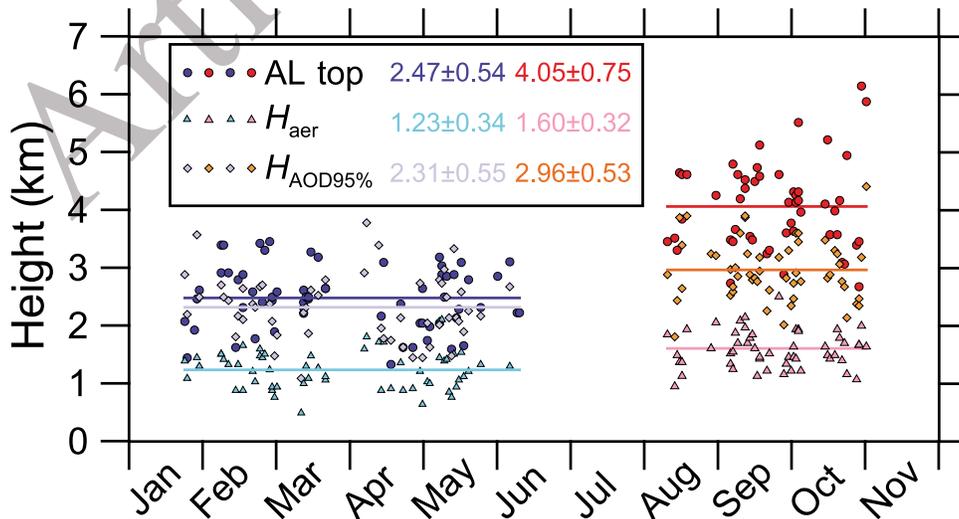


Figure 11. Time series of AL top, H_{AOD95} , and H_{aer} for the wet (blue colors) and dry season (red colors) in 2008. Horizontal lines indicate the mean values (given also as numbers).

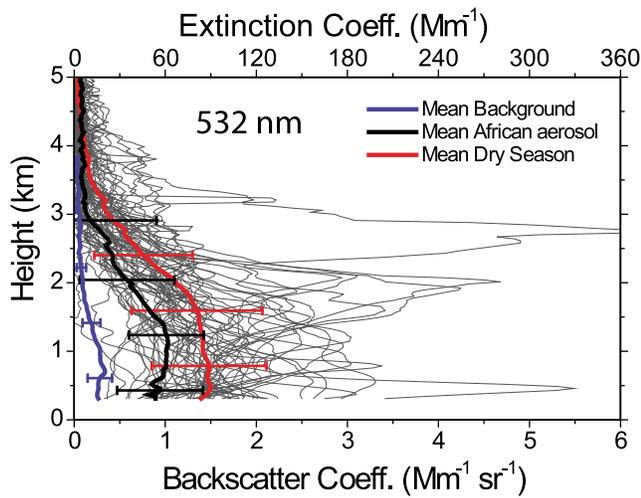


Figure 12. Mean profiles (thick solid lines) with standard deviations (thick bars, indicating the atmospheric variability) of the 532 nm particle backscatter coefficient for background conditions in the wet season (blue), conditions with African aerosol advection during the wet season (black), and as observed during the dry season (red). All individual profiles (thin grey lines) measured during the dry season are shown in addition to illustrate the strong variability and vertical inhomogeneity of smoke contamination during the burning season. Extinction scale (upper axis) is simply given by multiplying the backscatter scale with a lidar ratio of 60 sr.

675 between 1–1.5 for both, the short-wavelength and the long-
 676 wavelength range, which may be interpreted as a clear
 677 indication for the dominance of a well pronounced accu-
 678 mulation size mode of aged smoke particles. Similar dis-
 679 tributions were observed for aged Asian particles over the
 680 tropical Indian Ocean [Franke *et al.*, 2003] and for African
 681 smoke [Tesche *et al.*, 2011]. For the wet season, the broad
 682 Ångström exponent spectrum (Figure 13) for the 355–
 683 532 nm spectral range indicates less well defined aerosol
 684 conditions as a consequence of the occurrence of local
 685 aerosols (of biogenic origin with a small content of marine
 686 particles) as well as of African aerosols with variable frac-
 687 tions of dust and smoke.

688 **4.3.2. Extinction Profiling**

689 [55] Figure 14 shows the seasonal mean profile of the
 690 532 nm particle extinction profile for the dry season together
 691 with the individual profiles for more than 50 nights. These
 692 profiles are directly derived from the nitrogen Raman signals
 693 after cloud-screening. The profile of the corresponding mean
 694 Ångström exponent in Figure 14 is computed from the indi-
 695 vidual profiles of the Ångström exponent, which in turn
 696 are calculated by using the individual observations of the
 697 355 and 532 nm extinction profiles. A strong variability of
 698 the extinction values is found. Compared to the backscatter
 699 coefficients, which can be determined with high vertical
 700 resolution of 60–300 m, the Raman signal profiles are
 701 smoothed for the retrieval of the extinction coefficients with a
 702 vertical window length of 750 m. For this reason, the vertical
 703 variability of the extinction coefficient is reduced compared
 704 to the backscatter variability in Figure 12. On average during
 705 the dry season, 532 nm extinction values are mostly found

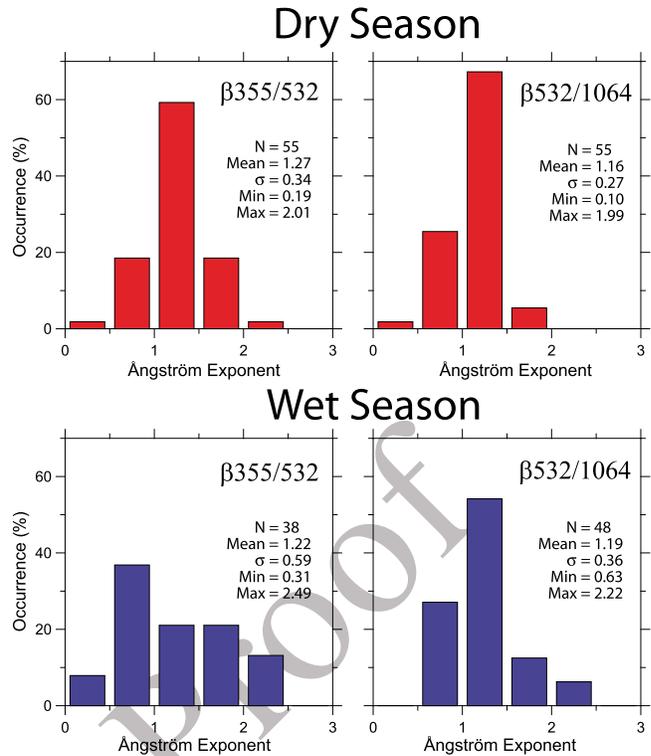


Figure 13. Frequency distributions of backscatter-related Ångström exponents for the two different spectral ranges from 355–532 and 532–1064 nm. The histograms are based on *N* layer mean values (1500–2500 m height range) for the dry (red) and wet (blue) season. Mean values, standard deviations, and maximum and minimum values are given as numbers.

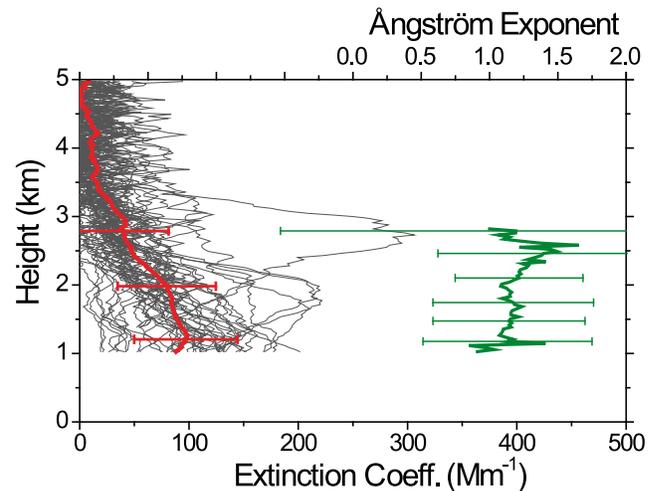


Figure 14. Mean profiles (thick solid lines) with standard deviations (thick bars, indicating the atmospheric variability) of the 532 nm particle extinction coefficient (red) and the Ångström exponent (green, 355–532 nm spectral range). These data are derived by applying the Raman lidar technique to the dry season observations. All individual extinction profiles measured during the dry season are shown in addition in grey to illustrate the variability and vertical inhomogeneity of smoke contamination.

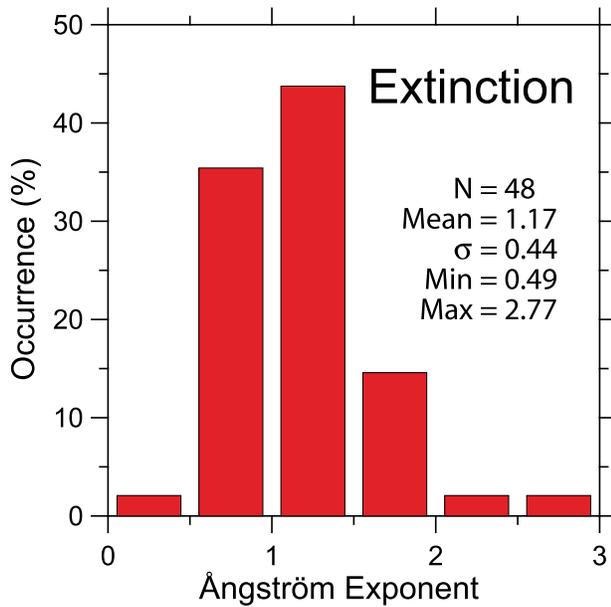


Figure 15. Frequency distributions of layer mean extinction-related Ångström exponent derived from N observations in the dry season. Mean values for layer from 1500–2500 m agl are considered. The statistics are based on N observations. Mean values, standard deviations, maximum and minimum values are given as numbers.

706 around 100 Mm^{-1} in the lowermost 2 km of the moist
707 atmosphere over the Amazon rain forest close to Manaus.

708 [56] As mentioned, wet-season extinction profiles data (by
709 using the Raman lidar method) can not be presented because
710 of the high frequency of low-level clouds, rain, and fog.
711 These unfavorable conditions prohibited temporal averaging
712 of the lidar signal for more than one hour which is necessary
713 for the determination of the extinction coefficient profile
714 with Polly^{XT} under very clean conditions.

715 [57] The mean profile of the extinction-related Ångström
716 exponent in Figure 14 is in agreement with the backscatter-

related Ångström values in Figure 13. On average, the values
717 range from 1–1.5 throughout the aerosol column. The fre-
718 quency distribution of the extinction-related Ångström
719 exponents found during the dry season is shown in Figure 15.
720 Frequently occurring values from 0.5–1.5 indicate compar-
721 ably large particles. The size of the particles may be widely
722 controlled by hygroscopic growth and growth during long-
723 range transport by condensation of gases and by coagulation.
724

[58] Similar Ångström exponent values were observed by
725 Schafer *et al.* [2008] for the northern Amazon rain forest
726 regions from AERONET observations. In the southern forest
727 and cerrado regions, however, the authors report a frequent
728 occurrence of Ångström exponent >1.5 during the dry sea-
729 son. Guyon *et al.* [2003] even reported Ångström exponents
730 above 2 over Alta Floresta in southern Amazonia. The
731 extinction-related Ångström values are in good agreement
732 with respective values found from Raman lidar observations
733 of aged biomass-burning plumes during the dry season in
734 southern, tropical Asia [Franke *et al.*, 2003] and during the
735 dry season in western Africa [Tesche *et al.*, 2011].
736

4.3.3. Extinction-to-Backscatter Ratio

[59] Figure 16 shows the lidar ratio (extinction-to-back-
738 scatter ratio) statistics for the dry season 2008. Mean values
739 for the central part of the smoke plumes (1500–2500 m
740 height) were analyzed. Lidar ratios ranged from 25 and
741 95 sr. The predominance of the lidar ratios were between
742 50 and 70 sr (in 61% out of all cases) for 355 nm and 50 and
743 80 sr (in 71% of all cases) for 532 nm. Together with the fact
744 that the Ångström exponents indicate, on average, relatively
745 large particle these high values indicate moderately to
746 strongly absorbing particles. For given chemical properties,
747 the lidar ratio decreases with increasing particle size and
748 decreasing absorption efficiency of the particle ensemble
749 [Müller *et al.*, 2007a]. In about 42% out of the cases, the
750 lidar ratios at 532 nm exceeded 70 sr which is clear sign for
751 strongly absorbing smoke particles. Such high values were
752 found for African smoke [Tesche *et al.*, 2011] and southern
753 Asian smoke [Franke *et al.*, 2003] originating from northern
754 India.
755

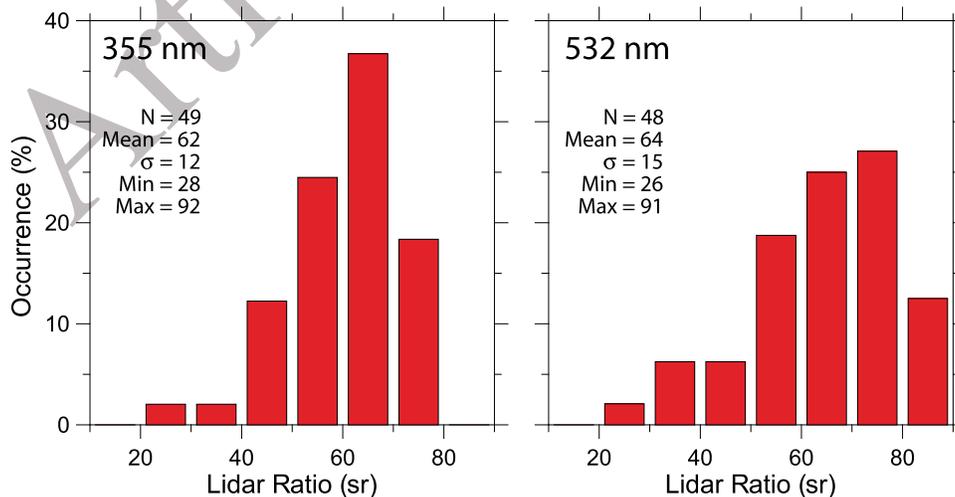


Figure 16. Frequency distributions of the layer mean lidar ratio at (left) 355 and (right) 532 nm. Lidar ratios for the layer from 1500–2500 m height are considered. Statistics are based on N observations. Mean values, standard deviations, maximum and minimum values are given as numbers.

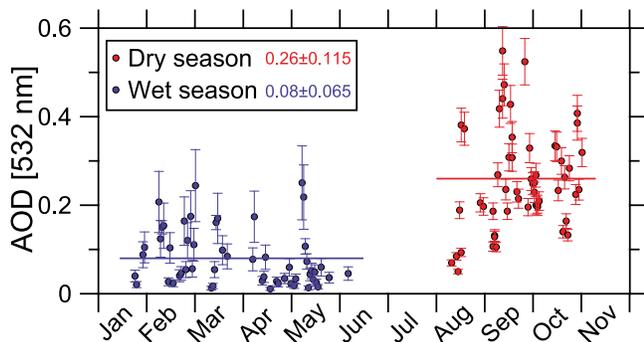


Figure 17. Time series of the 532 nm AOD during the wet season (blue) and the dry season (red) in 2008. Horizontal lines indicate the AOD means (given also as numbers). Horizontal lines indicate the mean values (given also as numbers).

756 [60] A unique feature of biomass burning aerosol is also
757 the wide spread of lidar-ratio values, here from 25–95 sr, as
758 mentioned above. This reflects the influence of particle
759 aging, including cloud processing of the particles, and
760 differences in the burning material and fire type.

761 **4.3.4. Aerosol Optical Depth**

762 [61] The time series of the 532 nm AOD measured with
763 the Polly^{XT} lidar from 22 January to 11 November 2008 is
764 shown in Figure 17. Pristine, clean background conditions
765 with AOD < 0.05 were often interrupted (on an almost regu-
766 lar basis) by advection of African aerosols causing AOD
767 values > 0.05. The cleanest conditions were most frequently
768 found in April and May 2008 because of the decreasing fire
769 activity at the end of western African fire season. The AOD
770 values did not exceed 0.25 during the entire wet season

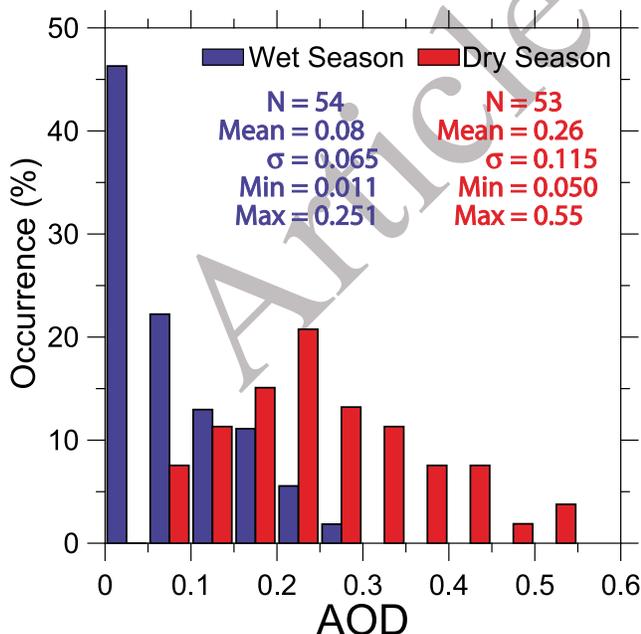


Figure 18. Frequency distribution of the lidar-derived 532 nm AOD for the wet (blue) and dry (red) season 2008. *N* observations were analyzed. Mean values, standard deviations, maximum and minimum values are given as numbers.

2008. The mean wet-season AOD was found to be 0.08 and
771 was thus about three times smaller than the one for the dry
772 season. However, under pristine background conditions a
773 mean AOD as low as 0.03 was observed whereas an average
774 value of 0.14 was found during periods with African aerosol.
775

[62] In the dry season, a high AOD variability was
776 observed. AOD values ranged from 0.05–0.55. The highest
777 AOD values were observed in September and October, when
778 the fire activity east and thus upwind of the lidar site was
779 highest.
780

[63] The observed inter-seasonal and in-season behavior
781 of the lidar-derived optical aerosol properties is in good
782 agreement with findings from measurements of optical
783 aerosol properties in Alta Floresta, Rondônia, in southern
784 Amazonia [Guyon *et al.*, 2003]. A high day-to-day variability
785 during the dry season was observed as well as a strong
786 contrast between the wet and the dry season in southern
787 Amazonia. However, the magnitude of the observed varia-
788 tions in the optical aerosol properties was much lower at the
789 lidar site. The area north of Manaus is obviously less affected
790 by smoke pollution than regions in southern Amazonia.
791

[64] The frequency distributions of the lidar-derived AOD
792 (532 nm) for the wet and dry season are shown in Figure 18.
793 Again, a strong contrast between the wet and dry season is
794 visible. Almost 50% out of all wet season observations yield
795 AODs < 0.05, whereas 50% out of the dry season cases
796 showed AODs > 0.25 at 532 nm.
797

[65] The ratio of AOD_{ML} of the mixing layer to the total
798 AOD for the dry season is shown in Figure 19 and indicates
799 that the mixing layer height can not be regarded as a barrier
800 for vertical aerosol exchange. In only 25% out of all cases
801 the AOD is controlled by aerosols in the mixing layer. These
802

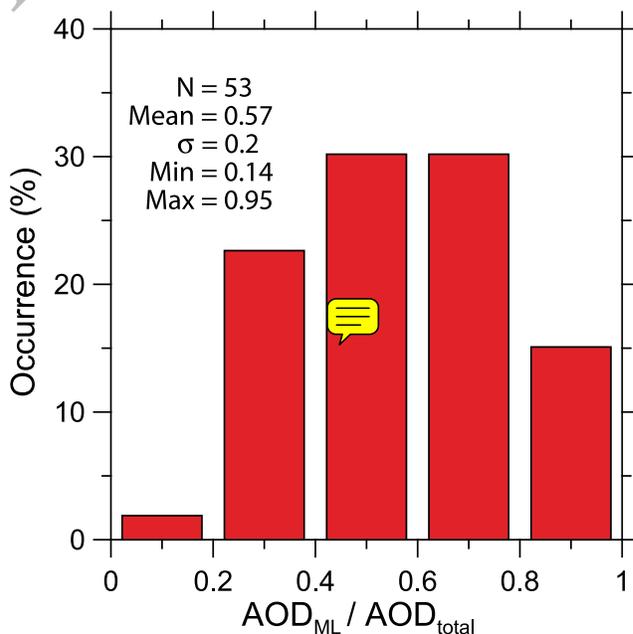


Figure 19. Frequency distribution of the AOD fraction that is contributed by the mixing layer (with top usually below 1500 m height) to the total tropospheric AOD for the dry season 2008. *N* observations were analyzed. Mean values, standard deviations, maximum and minimum values are given as numbers.

t2.1 **Table 2.** Monthly Mean AOD at 532 nm Derived From Lidar
 t2.2 Observations in 2008 and From AERONET Observations in the
 t2.3 Northern Forest Region for the 1999–2006 Period as Published
 t2.4 by *Schafer et al.* [2008]^a

		Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
t2.6	Lidar 2008	0.06	0.10	0.09	0.05	0.07	–	–	0.18	0.29	0.25	–	–
t2.7	AERONET	0.14	0.11	0.12	0.09	0.10	0.08	0.10	0.18	0.26	0.37	0.36	0.33

t2.8 ^aThe AERONET AODs were converted from 440 nm to 532 nm by using
 t2.9 the published monthly mean Ångström exponents.

803 findings corroborate results from airborne measurements
 804 during the dry season in Rondônia [*Guyon et al.*, 2005]. No
 805 differences in the aerosol characteristics within and above
 806 the mixing layer were found during these research flights.
 807 Our findings also agree well with airborne lidar observations
 808 by *Browell et al.* [1988] drawn from 11 flights in northern
 809 Brazil near the Amazon river in the summer of 1985. It was
 810 found that significant amounts of aerosol were above the
 811 mixing layer. The vertical distribution was generally very
 812 inhomogeneous.

813 [66] *Schafer et al.* [2008] analyzed several years of
 814 AERONET observations at 15 sites in Amazonia between
 815 1999 and 2006. Monthly mean AOD values at 440 nm were
 816 presented for the cerrado region, southern forest region, and
 817 the northern forest regions (the region of the EUCAARI
 818 lidar site). A comparison of monthly mean 532 nm AOD
 819 values determined with lidar and obtained from the AERO-
 820 NET photometers in northern Amazonia (4 stations, one
 821 close to the lidar site, 3 far more east) is shown in Table 2. A
 822 good agreement is found for the dry season.

823 [67] For the wet season, *Schafer et al.* [2008] reported
 824 AOD values between 0.1 and 0.14 (532 nm) in the northern
 825 forest region. This is much higher than observed with the
 826 lidar (0.05–0.1 at 532 nm). A stronger influence of African
 827 aerosol transport on the stations east of the lidar site may
 828 have caused this result as well as generally increased aerosol
 829 transport from Africa toward Amazonia between 1999 and
 830 2006.

831 [68] Finally, a summarizing overview of the lidar obser-
 832 vations during the wet and dry season 2008 is presented in
 833 Table 3. Mean values and respective standard deviations for
 834 the aerosol properties discussed before are given.

835 5. Summary

836 [69] For the first time the aerosol conditions over the
 837 Amazon Basin were continuously monitored in terms of
 838 height profiles of particle optical and microphysical proper-
 839 ties over almost one year. An automated, advanced Raman
 840 lidar was used that permits aerosol profiling at natural
 841 environmental conditions, i.e., at ambient humidity condi-
 842 tions (without any manipulation of the aerosol to be mea-
 843 sured as is needed in the case of in situ sampling).

844 [70] One of the key issues was to contrast the aerosol
 845 conditions during the wet season and during the highly
 846 polluted dry season. It was found that pristine environmental
 847 conditions occurring during the wet season were frequently
 848 interrupted by advection of African smoke and dust plumes.
 849 During these events, particle extinction coefficients and
 850 optical depths were observed to be enhanced by a factor of 4
 851 (on average).

[71] In the dry season, the backscatter, extinction, and
 optical depth values were, on average, increased by a factor
 of 6 in the main biomass burning layers at heights <2 km
 (compared to the pristine wet season aerosol conditions).

[72] Based on two cases studies it was shown that the
 optical properties of the biomass burning aerosol can be
 rather different as a result of the burning type, burning
 material, transport times, aging processes, and other effects.
 The findings confirm previous studies.

[73] The statistical analysis of the complete lidar data set
 revealed also strong differences between the pristine wet and
 the polluted dry season. Under pristine conditions, the par-
 ticle extinction coefficients at 532 nm wavelength were
 frequently as low as 10–30 Mm⁻¹, the particle optical depth
 was <0.05, and aerosol was trapped in the lowermost 2.5 km
 of the troposphere. During the dry season, the biomass
 burning smoke plumes reached to 3–5 km height. The AOD
 scale height was however usually below 2 km height. On
 average, particle extinction coefficients at 532 nm wave-
 length were of the order of 100 Mm⁻¹ in the main pollution
 layer (up to 2 km height) and 30–50 Mm⁻¹ from 2–4 km
 height. Ångström exponents were mainly between 1.0 and
 1.5, and lidar ratios accumulated from 50–80 sr. On average
 during the wet season 2008, the AOD at 532 nm was 0.03
 under background conditions and 0.14 during periods of
 African aerosol intrusion. A mean AOD of 0.26 was found
 during the dry season 2008.

[74] As already demonstrated in many aerosol lidar
 studies, the lidar ratios (especially when measured at two
 wavelengths) and the backscatter- and extinction-related
 Ångström exponents provide a solid basis for aerosol typing.
 Depolarization ratio observations are in addition of value in
 areas close to deserts or in outflow regimes of desert dust.

t3.1 **Table 3.** Mean Values and Standard Deviations of Lidar-Derived
 t3.2 Particle Optical Properties and Layer Height Parameter^a

Quantity	Dry Season	Wet Season	
<i>Tropospheric Column</i>			
AOD (355 nm)	0.38 ± 0.18	0.13 ± 0.06	t3.5
AOD (532 nm)	0.26 ± 0.12	0.08 ± 0.07	t3.6
AL top	4.1 ± 0.8 km	2.5 ± 0.5 km	t3.7
H _{aer}	1.6 ± 0.3 km	1.2 ± 0.3 km	t3.8
H _{AOD95}	3.0 ± 0.5 km	2.3 ± 0.6 km	t3.9
ML top	1.5 ± 0.4 km	1.1 ± 0.3 km	t3.10
<i>Mixing Layer</i>			t3.11
β (355 nm)	2.63 ± 0.99 Mm ⁻¹ sr ⁻¹	1.18 ± 0.52 Mm ⁻¹ sr ⁻¹	t3.12
β (532 nm)	1.44 ± 0.69 Mm ⁻¹ sr ⁻¹	0.62 ± 0.50 Mm ⁻¹ sr ⁻¹	t3.13
β (1064 nm)	0.50 ± 0.21 Mm ⁻¹ sr ⁻¹	0.24 ± 0.26 Mm ⁻¹ sr ⁻¹	t3.14
<i>1500–2500 m Height Range</i>			t3.15
α (355 nm)	118 ± 64 Mm ⁻¹	-	t3.16
α (532 nm)	70 ± 38 Mm ⁻¹	-	t3.17
β (355 nm)	1.83 ± 1.15 Mm ⁻¹ sr ⁻¹	0.41 ± 0.33 Mm ⁻¹ sr ⁻¹	t3.18
β (532 nm)	1.09 ± 0.67 Mm ⁻¹ sr ⁻¹	0.30 ± 0.35 Mm ⁻¹ sr ⁻¹	t3.19
β (1064 nm)	0.48 ± 0.20 Mm ⁻¹ sr ⁻¹	0.13 ± 0.18 Mm ⁻¹ sr ⁻¹	t3.20
Lidar ratio (355 nm)	62 ± 12 sr	-	t3.21
Lidar ratio (532 nm)	64 ± 15 sr	-	t3.22
a _{α355/532}	1.17 ± 0.44	-	t3.23
a _{β355/532}	1.27 ± 0.34	1.22 ± 0.59	t3.24
a _{β532/1064}	1.16 ± 0.27	1.19 ± 0.36	t3.25

^aValues are presented for the tropospheric column, for the mixing layer,
 and the central part of the smoke plumes (1500–2500 m height range).
 Particle backscatter coefficient, extinction coefficient, Ångström exponent,
 and particle optical depth are denoted as β, α, ā, and AOD, respectively.

885 The present study adds a new important data set to the lidar-
886 based global aerosol climatology. The study is simulta-
887 neously another example how powerful nowadays aerosol
888 lidars can contribute to atmospheric science related to atmo-
889 spheric composition and climate change. With automated
890 lidar systems this research can be done at even rather
891 inconvenient places like tropical forest (high humidity, high
892 concentration of insects that affect the optics and the overall
893 performance of the lidar).

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