First, we would like to thank the anonymous referees for their constructive comments and suggestions, helping to improve the manuscript. All referees remarked that the original manuscript contained too many figures. To comply with that, we reduced the number of figures from 21 to 14, by merging some plots and by removing others that were not crucial to support the data discussion. We replied to the reviewers' comments point by point, as follows.

### Anonymous Referee #1

1. Weather patterns play a major role in determining the annual or seasonal variabilities of aerosol optical properties at the site. I suggest more information of meteorological processes and their relationship to aerosol properties need to be added in the discussion. I am not convinced by the trajectory analysis presented in the text.

We agree that knowledge of meteorological processes can contribute significantly to the interpretation of aerosol data. We revised all through the text aiming to improve the discussion of how meteorological processes could have affected the observed aerosol optical properties. For example, we accounted for the effect of precipitation seasonality on aerosol optical properties, stating that the decreased particle scattering and absorption coefficients in the wet season resulted from both source strength increase and precipitation rate increase (refer to comment #11 by referee #2). We also discussed about the relationship between wind direction and aerosol properties. We recognize that it would have been better to run a regional atmospheric circulation model instead of using Hysplit back trajectories, particularly for relatively short distances like Manaus city - forest reservation. We also stated in the discussion section that in cloud aerosol processes may have significant effects on particle physical and chemical processes, particularly concerning to smoke aging processes. Unfortunately, observations of vertical wind velocity and vertical profiles of meteorological parameters were not available at the measurement tower, which restricts quantitative analysis of the relationships between aerosol optical properties, downdrafts and updrafts driven by convective clouds. In the manuscript we added a recommendation to improve observation of micrometeorological variables concomitant with aerosol measurements in future experiments. Finally, we think that the effect of precipitation scavenging and cloud processing on particle size distributions is a very interesting subject, and could be the theme of another paper, focused on case studies. We added this observation to the conclusion section.

- P23338 Line 14: Does "39 m" mean 39 m a. s. l. or 39 m above ground? It means 39 m above ground level, and this information was added to the text.
- 3. P23338 Line 22: What were the frequencies of full calibration and zero check of Nephelometer? It is better to mention this in the manuscript.

We agree that this information should be in the manuscript. We rephrased the sentence as follows: "Each six months the instrument was calibrated ..."

4. P23340 Line 27: It is not correct to use the temperature measured inside TSI Nephelometer to correct MAAP data. This temperature should be higher than the sample temperature because of the heating effect of the lamp.

As mentioned on line21 in the page 23339, the MAAP operated in series with the Nephelometer. Based on the sample flows (16.7 lpm) and on the dimensions of the tube connecting the instruments (0.432 ID and 15 cm), we estimate a sample traveling time in the order of 0.01 s. We consider that this traveling time is not enough for a significant temperature decrease in the way between the Nephelometer and the MAAP, and, therefore, we sustain that the temperature measured inside the Nephelometer is the best estimative for the sample temperature inside MAAP.

5. P23343 Line 14: As for the hemispheric backscatter ratio, new study shows that it is highly depend on the aerosol mixing state. Better to mention the impacts of mixing state on hemispheric backscatter ration here. See the following paper for more information. (Ma, N., Zhao, C. S., Müller, T., Cheng, Y. F., Liu, P. F., Deng, Z. Z., Xu, W. Y., Ran, L., Nekat, B., van Pinxteren, D., Gnauk, T., Müller, K., Herrmann, H., Yan, P., Zhou, X. J., and Wiedensohler, A.: A new method to determine the mixing state of light absorbing carbonaceous using the measured aerosol optical properties and number size distributions, Atmos. Chem. Phys., 12, 2381-2397, 2012.)

We are thankful for the reference suggestion, and added the following sentence to the manuscript: "Moreover, a recent study by Ma et al. (2012) have shown a significant dependency of the backscatter ratio on the aerosol mixing state."

- 6. P23345 Line 4: "has been investigated by (Kotchenruther and Hobbs, 1998)" should be revised as "has been investigated by Kotchenruther and Hobbs (1998)" Corrected, following the referee indication.
- 7. *P23346 Line 7: 1.59 should be 1.59-0i* Corrected, following the referee indication.
- 8. P23346 Line 25: The description of how to estimate the refractive indices is not clear. Does "calculated scattering coefficient" here mean the one which truncation and illumination function is considered in the calculation?

Yes, by "calculated scattering coefficient" we mean the Mie code calculated scattering coefficient taking into account the TSI Nephelometer truncation error (7-170°) and its non-Lambertian illumination function (Anderson & Ogren, 1998). In order to clarify this point, we rephrased the sentence as follows: "The refractive index representative of each size distribution was iteratively determined by means of matching the Mie code simulation for the Nephelometer signal, which takes into account its truncation error and illumination function, and the measured scattering and absorption coefficients within 10 %."

9. Fig3 and fig4: There are too many figures in this manuscript and some similar plots can be merged into one plot. It is better to combine fig 3 and fig 4 into one figure, i.e. plot the median number concentration on the wind rose figure.

By merging Fig 3 and Fig4, we believe that valuable information would be lost. We think it is important for the reader to have access to the histograms of wind speed magnitude throughout the measurement period (Fig 3a), since this is important climatological information which has not been published elsewhere and also helps to interpret the aerosol measurements at this particular site. Moreover, by plotting the median aerosol number concentration at a single wind rose plot, we would lose information about the differences between wet and dry season, which is crucial for the interpretation of the results. To comply with the reduction of the number of figures, we propose remove Fig3b, and substitute by Fig4.

- 10. P23347 Line 15: "30 to 3 h" should be "30 min to 3 h"?Yes, that is what we meant. This was corrected in the manuscript.
- 11. Fig5 and fig6: These two figures can be combined as two subplots.We followed the referee suggestion, and combined Fig 5 and Fig6 as subplots of the same figure.
- 12. Fig10, fig11 and fig12: These figures can be merged into one figure with three subplots. Following the referee suggestion, the figures were combined as subplots of the same figure.
- 13. P23352 Line11: The cloud cover was above 0.9 in 72% of the wet season days (P23355, line22). Is it possible to produce so many sub-micron particles via photochemical process under such cloudy days?

There are several evidences of secondary aerosol formation via photochemical processes in Amazonia, in spite of cloudiness. Measurements of aerosol chemical composition using a high-resolution mass spectrometer at the same forest site show mass spectra that resemble those of secondary organic aerosol particles formed in environmental chamber from biogenic precursor gases (Chen et al., 2009). From the perspective of gaseous precursors, observations have shown that there is plenty of biogenic volatile organic compounds in the Amazon (e.g. Rizzo et al., 2010), and that fast isoprene oxidation occur in the region (Karl et al., 2009), favoring the formation of of oxygenated products that might partition to the particle phase. To provide a reference to the reader, we added Chen et al. (2009) to the mentioned sentence.

- 14. Fig15 and fig16: It will be clearer to merge fig 15 into fig 16, just like fig 2. We merged Fig 15 and Fig 16 into a single plot, following the referee suggestion.
- 15. P23353 Line 25: All optic measurements were done for PM7 aerosol. But only particle number size distribution (PNSD) in size range of 10-500 nm was used for the relationship analysis. I do not know whether a better or worse correlation will be yield if PNSD for 10nm-7um is used. Is it possible to evaluate the influence of ignoring the PNSD of 0.5-7 um in the period that PNSD of supermicron is available?

This issue was raised by the three referees, and we recognize that this part of the manuscript needs adjustments. We agree that this is not ideal to compare

submicrometer particle size distribution parameters with intrinsic optical properties of particles with diameters between 10 nm - 7  $\mu$ m, particularly because scattering Ångström exponents are more sensitive to particles with diameters between 0.5 and 0.8  $\mu$ m. Therefore, we decided to present relationships between Ångström exponents and particle size distributions corresponding to the entire observed range, based on SMPS and OPC data. The drawback of this choice is that complete size distributions are available for only 90 days in the dry season of 2009, when backscattering data was lacking. Therefore, it will not be possible to make comparisons between wet and dry seasons, as well as study the relationship between backscatter and particle size. Thus, we rewrote section 4.3 as follows:

"In the literature, the scattering Ångström exponent has been used as an indication of particle number size distribution. Particles with diameters around 0.1  $\mu$ m, usually associated with urban pollution and biomass burning, have a steeper spectral dependency for scattering (e.g., Schuster et al., 2006). Therefore, this parameter is expected to decrease as the aerosol particle diameter increases. Mie theory calculations performed by Collaud Coen et al. (2007) suggest that the Ångström exponent is more sensitive to particles with diameters between 0.5 and 0.8  $\mu$ m.

The relationships with particle size were investigated through the comparison with the following weighted mean diameters calculated from aerosol particle number size distribution measurements (10 nm - 7  $\mu$ m) taken between July and August 2009: the count mean diameter (CMD),

$$CMD = \frac{\sum D_{P_i} N_i}{N_{total}} \qquad (4)$$

the surface area mean diameter (SMD),

$$SMD = \frac{\sum D_{P_i} S_i}{S_{total}}$$
(5)

and the volume mean diameter (VMD),

$$VMD = \frac{\sum D_{P_i} V_i}{V_{total}}$$
(6)

where  $D_{P\nu}$ ,  $N_{\nu}$ ,  $S_{i}$ , and  $V_{i}$  represents respectively particle diameter, number concentration, surface area and volume of bin i;  $N_{tota\nu}$ ,  $S_{total}$ , and  $V_{total}$  represents the corresponding parameters integrated for the whole diameter range. Particle size distributions in the range 0.3–6 µm were measured based on a physical assumption that tends to underestimate particle sizes (refer to section 3.2). In this range, particle size spectra were corrected by means of optical closure study, using a Mie code to infer what size distribution would fit to the observed particle scattering and absorption coefficients (refer to section 4.5).



Figure shows that the correlations between Ångström exponents and weighted mean diameters are rather poor. Nevertheless, it shows that Ångström exponents decreased with SMD and VMD, as expected. The dependency of the Ångström exponent with CMD contradicted the expectations, increasing with particle size. This converse behavior has been reported for aerosols in a Chinese megacity (Garland et al., 2008) and in a boreal forest site (Virkkula et al., 2011), and might be related to the fact that surface and volume are more correlated to particle size than are number concentrations. Schuster et al. (2006) argue that for bimodal aerosol particle number size distributions the Ångström exponent can decrease with particle size, depending on the ratio of fine and coarse particle concentration. The addition of coarse mode particles with spectrally flat extinctions reduces the overall spectral variability, decreasing the Ångström exponent and damping its sensitivity to the size of fine mode particles."

16. P23356 Line 25: There are two kinds of model calculated scattering coefficients: the normal one and the one simulating Nephelometer. Also there are two measured scattering coefficients: the raw data and the corrected data. It is confusing in the text on which kind of measured and calculated scattering coefficients they are.

For the sake of data closure, the correction factors on scattering coefficients due to instrument non idealities were calculated using two approaches: Anderson and Ogren (1998) parameterization (section 2.2), and Mie modeling (section 4.5). We recognize that the terms used in the text might be confusing. Concerning to that, we revised the text throughout and rephrased a couple of sentences, as follows:

P23339 Line 1: "The average correction factor for truncation errors, <u>calculated as the</u> ratio between corrected and raw data, was 1.13±0.08 for scattering coefficients at 550

nm. As will be discussed on Sect. 4.5, this truncation correction factor fits well to Mie code correction factors, <u>taken as the ratio between the simulated integral scattering</u> <u>coefficient (0-180°) and the simulated Nephelometer signal considering its angular</u> <u>truncation (7-170°) and illumination function</u>."

P23356 Line 25: "<u>The modeled truncation error was calculated as the ratio between as</u> <u>the simulated integral scattering coefficient (0-180°) and the simulated Nephelometer</u> <u>signal considering its angular truncation (7-170°) and illumination function.</u> In average, the model predicted 15±5% underestimation of 550 nm particle scattering coefficients due to the Nephelometer truncation error."

17. P23357 Line 3: Are 0.07 and 0.005 the standard deviations or the estimated uncertainties of the calculated refractive index? Several factors can induce uncertainties to the calculated refractive index, such as the measurement uncertainties and the assumption of aerosol mixing state. Is it possible to evaluate the uncertainties of this value?

The numbers refer to the average and standard deviations of the calculated refractive index. As the referee mentioned, several factors contribute to the uncertainties associated with the calculated refractive index. We recognize that this should be clarified in the text, and we corrected it accordingly. In order to estimate the accuracy of the calculated refractive index, we varied the Mie model input data (from Nephelometer, MAAP, SMPS and OPC) by a quantity equal to the instruments error estimates ( $\pm \sigma$ ), mentioned on Section 2.2 This sensitivity test was applied for ten representative cases in the dataset. On average, the results showed variations of 4% on the real part of the refractive index, and of 15% on its imaginary part, as showed in the figures below (not to be included in the manuscript). Adding to the measurements uncertainties, there are errors related to the assumption of homogeneous internal mixture and sphericity throughout the whole measured size spectrum (10-7000 nm), which are difficult to quantify, and should be the subject of another paper. Therefore, the values obtained for the refractive index must be interpreted as an effective refractive index for the whole aerosol population.





#### Anonymous Referee #2

- Abstract:
  - 1. P. 23336, lines 1: the sentence leads to misunderstanding: the authors want probably to say that advection from Africa lead to an enhancement of crustal element in fine mode particles. The present sentence let thing that there is an enhancement of fine mode particles, what is not the case (Fig. 15).

We recognize the misunderstanding, due to a comma that was not supposed to be there. We rephrased as follows: "... characterized by enhanced concentrations of crustal elements (AI, Si, Ti, Fe) and potassium in the fine mode (PM2.0)."

- Introduction:
  - p.23336 line 14: "the region": please specify once again "the Amazon basin" since several other regions are cited in the previous sentence.
     We replaced "region" by "Amazon Basin", following the referee suggestion.
- Experimental:
  - p. 23338 line 4: please give the size of the reservation either in the text of in Fig. 1.
    The Cuieiras forest reserve encloses 380 km<sup>2</sup> of tropical forest, and this information was added to the text according to the referee suggestion.
  - 4. P. 23339 and Table 1: I am quite surprised to see an error at 700 nm two times smaller than the one at 550 and 450 nm for a scattering coefficient of 1 Mm-1.

We double checked the calculation of scattering coefficient errors, and confirmed the values displayed on Table 1. Indeed, the error at 700 nm is smaller than the error at 450 and 550nm for a scattering coefficient of 1 Mm<sup>-1</sup>. This is because the offset variability ( $\delta$ O) for 700 nm, as reported by Anderson & Ogren (1998) in their Table 2, is two times smaller in comparison to  $\delta$ O at 450 and 550 nm, making the calibration drift error smaller at this particular wavelength and scattering coefficient. The offset refers to the Nephelometer calibration with CO<sub>2</sub> and particle-free air. For the scattering coefficients of 10 and 100 Mm<sup>1</sup>, the smaller  $\delta$ O at 700 nm is compensated by the

increased slope variability ( $\delta$ S) and increased wall scatter (W) at 700 nm, according to Tables 2 and 3 of the referred article. This compensation results in similar errors at the three wavelengths when dealing with scattering coefficients above 10 Mm<sup>-1</sup>.

- 5. *P. 23339 line 16: backscattering instead of back scattering* Corrected, following the referee indication.
- 6. P. 23340 line 5-9: I find the MAAP error of 4% very small. I remember a previous estimate made by A. Petzold leading to 12% error. I wonder if the authors have taken into account all possible errors, since the present reported error is much less than all the errors for scatter and backscatter coefficients.

Indeed, the 4% uncertainty refer only to instrument noise and unit to unit variability, based on an intercomparison of filter based light absorption methods (Müller et al., 2011). The same study reports that the MAAP cross sensitivity to non-absorbing aerosol, i.e., scattering as apparent absorption, is on average 1% of the scattering coefficient. This percentage depends on the aerosol loading and single scattering albedo. Taking that into account, the overall MAAP uncertainty under typical Amazonian conditions is 10% (see table below – not to be included in the manuscript), in agreement with the reviewer comment. We corrected the manuscript accordingly: "A recent intercomparison of absorption photometers report MAAP noise levels up to 0.22 Mm<sup>-1</sup> for 1 min averaging time, unit to unit variability of 3% <u>and 1% of the scattering seen as apparent absorption, depending on the aerosol loading and single scattering albedo</u> (Müller et al., 2011). Assuming that the instrument noise decreases with the square root of averaging time, total uncertainties on aerosol absorption measurements averaged each 30 min <u>and under typical Amazonian conditions</u> is of 10%. "

$\sigma_{abs} [Mm^{-1}]$	σ <sub>scat</sub> [Mm⁻¹]	$\delta_{\text{noise}}$	$\delta_{unit\_variability}$	$\delta_{apparent\_scat}$	$\delta_{\text{total}}$	%
1	10	0.040	0.03	0.1	0.112	11%
10	100	0.040	0.3	1	1.04	10%
100	1000	0.040	3	10	10.4	10%

7. p. 23347 lines9-11: in the text or in the figure caption of Fig. 3, please specify the location (in degrees) of Manaus and the diesel generator or the wind direction bringing influenced air masses. For the diesel generator it is given in the following §! Please put the information before.

Following the suggestion, we rephrased the sentence as "... in which the signal of the Manaus urban plume ( $120-190^{\circ}$ ) and of the diesel generator ( $270-340^{\circ}$ ) is clear during the wet season."

8. Figure 4: wind direction is probably given in °.

Yes, wind direction is in degrees. We added the symbol  $^{\circ}$  to the x-axis label.

9. P. 23347 line17-18: please reformulate: it seems that it is the analysis that comprise 2% of the measurement period. Idem p. 23348 line1: "comprising" should be changed, for example by "corresponding to".

We accept the referee suggestion, and replaced "comprise" for "correspond to".

#### Results and discussion

10. p. 23349 line15-19: If the difference between 2009 and the other years is due to fires, it seems surprising that the scattering coefficient is much more (2 times) enhanced (110%) than the absorption coefficient (23%), since fires probably produced a lot of black carbon. Can you please explain this difference between scattering and absorption coefficient?

To elucidate this issue, we propose the addition of the following sentences (underlined) to the manuscript: "Error! Reference source not found. shows that the 2009 dry season had median values of aerosol particle scattering and absorption respectively 110% and 23% greater than the values observed during the dry season of 2010. This may be an outcome of the increased occurrence of fire spots at neighbor municipalities and at districts located to the East of the forest reserve during the dry season of 2009 in comparison to 2010 (Error! Reference source not found.). The same table shows that in 2010 there was profusion of fires, but in the Amazon region as a whole. The fact that median scattering coefficients were much more enhanced than absorption coefficients in 2009 can be surprising at first. However, during the dry season, the air masses reaching the measurements site constitute a blend of smoke plumes from different fire stages and ages. Reid et al. (2005) states that deforestation fires can smolder for days, producing particles at high emission factor rates with small black carbon content, thereby decreasing the aerosol absorption and increasing aerosol scattering of the original plume. Evidences show a tendency of particle scattering increase with plume age, due to physical and chemical atmospheric processes acting towards particle size increase, mass increase and gas-to-particle exchanges (Reid et al., 2005 and references therein). Conversely, in situ observations of biomass burning plumes indicate that black carbon concentrations typically decrease by only 10-50% from fresh smoke to regional haze (Capes et al., 2008; Reid et al., 1998), through dilution with cleaner background air. Therefore, this is reasonable that particle scattering and absorption coefficients vary by different factors, depending on the plume characteristics."

11. p. 23349 lines 20-22: The higher scattering coefficient during dry season isn't also (and principally) a consequence of a larger aerosol concentration due to a lower removal by precipitation?

Between 2008 and 2011, from dry to wet season, particle number concentrations (Dp>10 nm) increased by a factor of 3, fine mode particle mass (Dp < 2  $\mu$ m) increased by a factor of 2, and average daily precipitation decreased by a factor of 2 (from 10 to 4 mm/day). Having in mind the global rainfall climatology (e.g., Kidd, 2001), a precipitation rate of 4 mm/day is not insignificant. Anyway, we agree that the enhanced particle scattering coefficients in the dry season result both from increased

particle loading and reduced rainfall. We propose to add the following sentence to clarify this issue in the manuscript: "Lower removal by precipitation (from wet do dry season the average precipitation rates decreased from 10 to 4 mm.day<sup>-1</sup>) may also play a role on particle scattering increase in the dry season."

12. P.23350 lines 6-11 and Fig. 7 and 8: The absorption coefficient is enhanced in January and February, as well as in a lower extend in March 2010. The PM2 crustal elements are enhanced to a much lower extend in January and February 2010, but to a larger extend in March 2010. The 2008 February to May period present however a high concentration of crustal elements that is not mentioned and explained in the text and that has no correlation with the absorption coefficient (Fig7). Potassium has a higher proportion in January and February 2010, but the potassium in the fine mode is associated with to biomass burning and biogenic sources in the Amazon in the text (lines 8-10). How can then all these results convince the reader that the increase in absorption in January-February 2010 relates to advection of mineral dust and biomass burning from Africa??? This isn't coherent.

We thank the referee for the observation, since it helped us to fix some distortions regarding to Figures 7 and 8, and also to improve the discussion of the results. First, we may highlight that the integration time of the aerosol filter samples varied between 3 to 6 days, while the MAAP integration time was 30 min. Hence, we reasoned that monthly statistics are not appropriate, either because some of the filter integration times fallen between the end of a month and the beginning of the next month, and because the aerosol absorption data during African advection episodes might be obscured in monthly statistics due to the relatively high measurement frequency of absorption (30 min) combined with the duration of the events (2-6 days). Instead of monthly statistics, we decided to show aerosol absorption averages according to the filter integration times. Another distortion on Figures 7 and 8 is that we were comparing absorption (an extensive aerosol property) to fine mode relative percentages of crustal elements and K. To fix that, we decided to show absolute mass concentrations instead of relative percentages. The result is shown in the new Figure 5 (all updated figures are shown after the referee's questions), by which we intend to substitute Figures 7 and 8. Accordingly, we propose to rewrite the paragraph between Page 23349 (lines 23-26) and Page 23350 (lines 1-11) as follows:

"Another feature shown in **Error! Reference source not found.** is that the median value of the particle absorption in the wet season of 2010 was approximately 2.6 times greater than the values observed during the other years of measurements. Between January and February 2010, eight periods of elevated absorption coefficients (> 2 Mm<sup>-1</sup>) were observed, lasting continuously from 2 to 6 days. No influence of the Manaus urban plume or fire spots in nearby districts was detected in this period. This time scale is typical of African aerosol advection events, and this is a possible explanation for the observed increase of absorption coefficients in the wet season of 2010. Figure 5 shows particle absorption coefficients averaged according to aerosol filter integration times (2-6 days), as well as fine mode crustal elements concentration (Al, Si, Ti, Fe) and fine mode potassium concentration. Potassium in the fine mode has been associated both to biomass burning and to biogenic sources in the Amazon (Artaxo et al., 1994). For simplicity, in Figure 5 only the data between Jan and May of

each year is shown, since this is the period when most African advection events occur. The February 2008 African advection event (refer to **Error! Reference source not found.**) is clearly depicted, with increased concentrations of fine mode crustal elements and potassium, as well as increased particle absorption. The April 2008 advection event had a clear effect over the aerosol filter samples, but not over the particle absorption coefficients. The reason for that is unknown, but could be related to the fact that occasionally mineral dust particles are internally mixed with organic material, possibly affecting the aerosol optical properties (Pöschl et al., 2010). Between January and February 2010, Figure 5 shows enhancements of potassium in the fine mode associated with particle absorption. This could be explained by African advection of aged biomass burning particles, but the possibility of biogenic aerosols affecting the absorption coefficients cannot be ruled out. In accordance with that, Guyon et al. (2004) estimated that 35% of light absorption could be attributed to biogenic particles in another Amazonian forest site. In March 2010 the absorption data coverage was very poor (14%)."

13. P. 23350 lines17-19: the increase of the column AOD due to African advections that is not detected by the in-situ measured extinction is not clearly visible on Fig. 9. Some cases can be identified in Jan-march 2011 (large AOD aeronet), but similar cases can be found during the dry season without Africa advection (for ex. Dry period 2008). Perhaps another representation could allow to see the described effect.

The observation that AOD sometimes increases without a corresponding enhancement on in situ aerosol extinction also holds for smoke particles advected from inside Basin areas in the dry season. Since there is a lack of space for new figures and tables, we intend to keep Figure 9 as it is, and just rephrase the sentence as follows:

"It is worth noticing that sometimes the remotely sensed AOD increased without a corresponding enhancement on in situ aerosol extinction. This is an indication that not all advected aerosol plumes, originated either at Africa or inside the Amazon Basin, reached the surface at the measurement site. "

14. P. 23351 lines 10-12 and fig. 13: the aeronet retrieved SSA is given for the 1993-2011 period is compared to the in-situ 2008-2011 SSA. It would be much better to have the same averaging periods to compare both SSA. Various trends and particular events could have happened in the 15 years that are not taken into account in the in-situ measurements.

We agree that it would be better to have the same averaging periods and same locations to compare SSA from AERONET and from in situ aerosol measurements. Nevertheless, AERONET SSA measurements are very scarce in the Amazon Basin, either because of the low aerosol optical thickness in the wet season (<0.4) and because of the steady cloud cover, particularly over forest land. If we consider only the AERONET data between 2008 and 2011, we would have 442 SSA records for the arc of deforestation region, heavily impacted by biomass burning emissions, and only 9 SSA records from the Manaus AERONET station, in northern Amazonia. We considered that 9 data points in three years may not be enough to represent the Amazon Basin pristine areas, also because these measurements are in the confidence level of 1.5. Therefore,

to improve statistics and provide means for comparisons, we decided to include all SSA data ever observed in the Amazon Basin.

15. P. 23351 lines 18-23: is this information in the scope of this paper?

We believe that this is important to mention the number of available AERONET SSA data records, to give the reader a sense of how scarce is this kind of measurement in the Amazon Basin. It is remarkable that in 18 years of AERONET measurements, only 129 SSA data records could be retrieved for the Northern Amazonia, where sits most of the untouched forest land areas.

16. P. 23352 line 5-9: Fig. 14 does not allow to see an increase of 50% of the scattering coefficient (6-5 to 10 Mm-1). It has to be mentioned that the assessment of the second sentence is not shown.

To be precise, the median scattering increase between 7:00 and 15:00 in the wet season was of 54% (from 6.5 to 10 Mm<sup>-1</sup>), while the median absorption increase was of 53% (from 0.38 to 0.58 Mm<sup>-1</sup>). If the referee senses that this is hard to see the 54% scattering coefficient increase on Figure 14a due to its reduced size, we propose to remove the subplots 14b and 14d, for the sake of clearness. These subplots refer to the dry season, which did not show a clear diurnal cycle for scattering and absorption coefficients, as mentioned in the manuscript.

Concerning to the second sentence, we rephrased it as follows: "In the wet season, sub micrometer aerosol particle number size distribution measurements (not shown) indicated an increase of 20% ..."

17. P. 23352 line 10: The daytime shift of sub micrometer particle diameters towards larger sizes...

"... towards larger sizes..." was added to the refereed sentence.

18. P. 23352 line 13-15: you observe a greater enhancement of the absorption than of the scattering coefficient. However your explanation given in lines 13-15 would induce the opposite, that is a larger diurnal cycle of the scattering than the one of the absorption coefficient! Please explain.

As stated two comments before, the median scattering increase was of 54% (from 6.5 to 10 Mm<sup>-1</sup>), while the median absorption increase was of 53% (from 0.38 to 0.58 Mm<sup>-1</sup>). Thus, we understand that we observed similar daytime enhancements of absorption and scattering during the wet season. Mie model calculations indicated that the mentioned 20% diurnal particle growth had a greater effect over scattering (50-70% increase) compared to absorption (10-40% increase). We recognize that the diurnal particle size increase may not explain the entire absorption enhancement, and propose to add the following the sentence to the manuscript: "While the particle growth may be enough to explain the diurnal particle scattering enhancement during

the wet season, there may be other factors, still unknown, contributing to the observed particle absorption diurnal increase.".

19. P. 23352 line 17 and figure caption 14: I didn't find the word "diel" in my dictionary. Do you mean "diurnal"?

"Diel" is the terminology adopted by Dr. M. O. Andreae for hour-of-the-day data patterns. The idea is to avoid "diurnal cycle" being misunderstood by day-only cycle. Nevertheless, we will follow the referee indication and replace "diel" per "diurnal".

20. P. 23353 lines 12-14: and how many % for the wet season?

To comply with the referee question, we added "and 1-2% in the wet season" to the mentioned sentence.

- 21. P. 23354 lines 14-15: this affirmation is quite difficult to verify with Fig. 17. Fig. 17 principally shows that:
  - o The correlation is rather poor between the compared variables
  - o The dry season leads to better results than the wet season
  - o The correlation is always better with VMD and SMD than for CMD, what can be easily explained by the fact that the Ångström exponent and the backscatter ratio are sensitive to the size (that is more correlated to the surface and the volume) and not to the number of particles
  - o The fitted lines correspond to nothing and cannot be used as eye guide
  - o It seems to have a somewhat better correlations with the Ångström exponent than with the backscatter ratio.

All this should be better described under 4.3! Moreover Fig. 17 is restricted to the 10-500 nm particle diameter whereas the Ångström exponent is explained in the text to be sensitive to aerosol between 0.5 to 0.8 micrometer. In the text (p. 23355 line 5) coarse aerosol are also mentioned.

Please refer to comment #15 by referee #1.

22. P. 23355 line 24: "below" means that the forcing efficiency is smaller (more negative) than -3.5, but it is between -3.5 and 0!

We agree, and rephrased as follows: "...the magnitude of the negative aerosol forcing efficiency was below  $3.5 \text{ W.m}^{-2} \dots$ "

23. P. 23355 line 26-27: the indirect effect is the modification of cloud albedo and cloud life time by the aerosol. It is not the presence of clouds. The author cannot therefore make the conclusion that the indirect aerosol effect is larger than the direct one.

We agree that the presence of clouds by itself do not imply that the aerosols have changed cloud albedo and life time. The only statement we can make is that "in the wet season the radiative balance is dominated by the cloud cover, or, in other words, the radiative aerosol direct effect may not be important.", and we removed the mention of the aerosol indirect effect. 24. P. 23355 line 27-p. 23356 line 1: where can it be seen that the direct effect can be as important than the indirect one in the dry season? Just because the cloud cover is of about 50% ' + see previous comment.

In accordance with the previous comment, we rephrased the sentence as follows: "In the dry season the radiative aerosol direct effect gains relevance, and can be as important as the cloud cover radiative effect, at least referring to aged biomass burning particles."

25. P. 23356 line 8-13: As it is clearly explained, the only reasonable comparison between the in-situ calculated forcing efficiency and the one measured by REM is for days without clouds. The authors have to do this comparison and not the one described in the paper.

As the manuscript states in lines 12-15, within the time period of backscattering in situ measurements there were only 10 days with cloud fraction less than 0.1, for which a comparison of in-situ and remote sensed aerosol forcing efficiency could be made. Only three out of ten low cloud fraction days occurred in the wet season. In spite of the poor statistics related to the reduced number of data points, the manuscript mentions in line 14 the average aerosol forcing efficiency of  $-46 \pm 9$  Wm<sup>-2</sup> for the wet season and of  $-51 \pm 10$  Wm<sup>-2</sup> for the dry season, considering only days with cloud cover less than 0.1. These values are higher than the ones reported by Sena (2012) (and references therein) for the 24-hour aerosol forcing efficiency averaged along 9 years of remote sense observations in Amazonia. Although, this is hard to compare 7 days in the dry season of 2010 to 9 years of remote sense observations. Also, it is worth to mention that the methods used for the aerosol forcing efficiency calculation are completely different: the method used for remote sensed data takes the forcing efficiency as the slope of the curve Flux at TOA versus AOD, usually at 550 nm, for a entire dry season data set; on the other hand, the method used for in situ data uses daily records of spectrally averaged backscatter and single scattering albedo, without any mention to the AOD wavelength. Therefore, a forcing efficiency value calculated from in situ observations corresponds to a single point in the curve Flux at TOA versus AOD, which in many cases have a scattered linear correlation. Thus, the comparison suggested by the referee is not straightforward, considering the available data set of in situ observations. The purpose of the 4.4 section in the manuscript is to estimate the aerosol forcing efficiency from in situ observations in a forest site in Amazonia, and to bring attention to the fact that clouds play a key role in the Amazonian atmospheric radiative balance, particularly over forested areas not impacted by fresh biomass burning smoke. After this reasoning, we propose to add the following sentences to the end of section 4.4:

"These values are 4 times higher in magnitude than the ones reported by Sena (2012), averaged along 10 years of remote sense observations in Amazonia. Nevertheless, it is worth to mention that the methods used for calculating the aerosol forcing efficiency from remote sense and in situ data are completely different: the former takes the forcing efficiency as the slope of the plot of radiative flux at the top of the atmosphere versus AOD, usually at 550 nm, for a entire dry season data set; on the other hand, the

latter uses daily records of spectrally averaged backscatter and single scattering albedo, without any mention to the AOD wavelength. Therefore, a forcing efficiency value calculated from in situ observations corresponds to a single data point in the curve radiative flux versus AOD, which in many cases shows a scattered linear correlation."

- *26. P. 23356 lines 13-15: compare these values with REM ones.* Answered in the previous topic.
- 27. P. 23357 line 3: please mention also here briefly how this refractive index was found. We rephrased the sentence as: "The average refractive index resulting from the optical closure study was ..."
- 28. P. 23359 line 10: 1-15 points over how many?We rephrased the sentence as: "... and the trajectories enclosed 1 to 15 points (out of 24) inside the Manaus area."
- 29. P. 23359 last §: redundancy, please remove as well as Fig. 21

We think that Figure 21 is illustrative of the Manaus plume effect over the natural environment. However, to comply with the need of reducing the number of figures, we propose to remove Figure 20 instead, since it shows the absence of clear relationships between aerosol properties and the age of the urban plume.

# • Conclusion:

30. This section should not only list the previous discussed results, but also synthesize them to obtain a global picture of all results.

The conclusion section was reformulated, complying with the referees' questioning on specific points, and the results were better synthesized. Also, we mentioned in this section the role of meteorological processes on aerosol properties, and the importance of measuring micrometeorological variables simultaneously to improve the aerosol data interpretation.

- Tables: there is globally a lot of tables!
  - 31. Tables 5: redundancy with Figures? Please use always quartiles or percentiles, but no mix of them.

Following the referee suggestion, we replaced "percentiles" by "quartiles" in Table 6 and its legend. If possible, we would like to keep Table 5 (or at least move it to supplementary material), because it supports the discussion on year to year variability of aerosol optical properties, and may be in a useful format for modelers.

# • Figures: there is also too much figures!

32. Fig. 2: % of data necessary?

We think that the percent data coverage is necessary here, to avoid misinterpretation. For example, the reader might interpret the absence of data in October 2008 as if no precipitation occurred in that month. *33. Fig 5-6: one figure?* Following the referee suggestion, we will merge Figures 5 and 6 into one.

- *34. Fig. 7-8: one figure?* Please refer to comment #12.
- 35. Fig. 8: labels not clear (once PM2 crustal and once k/PM2), please put a) and b) in the figure and figure caption Please refer to comment #12.
- 36. Fig. 9, not very readable

To account for that, we improved the contrast between the data points in the figure.

37. Fig 10- 11: redundancy with Fig 5-6 and tables? it is not clear that you make a mean 2008-2011 seasonal cycle and why do you use 10 days means . (idem for Fig 12)

While Figures 5 and 6 show the year to year variability, Figures 10 and 11 focus on seasonal variations. Therefore, we would like to keep Figures 10 and 11 if possible. The ten days statistics were arbitrarily chosen to improve the readability of the plots. Visually, it gets much polluted if one considers one day statistics instead. To make clear that the figures refer to a median seasonal cycle, we replaced "Box plot" by "Median 2008-2011 seasonal cycle" in the captions of the Figures 10-12.

- 38. Fig. 10-16 the dry season should also be colored in rosa as in previous Fig.We accept the suggestion, and will add red shades to the mentioned Figures, as long as it does not affect its readability.
- 39. Fig. 16: one fig with 2 axis

We accepted and implemented the suggestion.

- 40. Fig. 17: better 2x3 fig than 3x2 fig! + see comments in textFigure 17 was reformulated, according to the comment #15 by referee #1.
- 41. Fig 20-21: not necessary As explained before, we propose to keep Figure 21 and remove Figure 20.

# Anonymous Referee #3

Specific Comments:

- Abstract:
  - Pg. 23335, Line 2 and 18: Pristine? May reconsider the use of the word 'pristine'. On Pg. 23337, Lines 25-26, the authors state that this region 'is influenced by external aerosol sources like regional biomass burning, urban plumes and African dust advection.' And on Pg. 23338, Lines 9-10, they also state "... the site was affected by regional transport of pollutants, either from biomass burning or urban plumes." This is not pristine; it is an occasionally perturbed site.

We agree, and reconsidered the use of the word "pristine" along the text, beginning with the title: "Long term measurements of aerosol optical properties <u>in a primary</u> <u>forest site</u> in Amazonia"

- Line 5: 'major' classes of particles
  We accept the suggestion, and corrected the text accordingly.
- 3. Line 12: what is fine mode? Define here rather than on Pg. 23336, Line 1. We accept the suggestion, and corrected the text accordingly.
- 4. Line 16: replace 'particle' with 'aerosol'.We accept the suggestion, and corrected the text accordingly.
- Line 27: replace 'advection' with 'advected'.
  We accept the suggestion, and corrected the text accordingly.

### • Introduction and Discussion:

- 6. *Pg. 23337, Line 8: replace 'focus' with 'focuses'.* Corrected.
- 7. Pg. 23338, Lines 12-15: Has any study of the size-dependent particle passing efficiency of the inlet been performed? An inlet of this length would be expected to show losses for small particles by diffusion. If small particles are preferentially lost, how does this affect the results for SSA and for the SMPS measurements? Also, no mention is made of the sampling lines to the instruments. These lines may not have laminar flow and other loss processes (e.g., gravitational settling or inertial losses) may come into play. Can the authors please comment on this? How severe are the losses at the small and large ends of the size distribution?

A study of the size-dependent particle loss trough the inlet was performed along the AMAZE experiment (Martin et al., 2010) in 2008, at the same site and set up. For the dimensions and flow of the laminar sampling line, diffusional and gravitational deposition losses for a particle of dynamic shape factor of 1 and a density of 1000 kg.m<sup>-3</sup> were calculated, indicating 50% transmission cutoffs of 4 nm and 7 µm through the sampling lines, with increased transmission between those sizes. This was further confirmed by a good agreement between the particle number concentrations recorded using condensation particle counters (CPCs) inside the TT34 container and those recorded by a CPC on the top of tower K34, approximately 1 km away from the TT34 tower. More details can be found in Martin et al. (2010) paper. Moreover, even if there were severe losses for ultrafine particles are not efficient light scatters nor absorbers, as suggested by Figure 19 in our manuscript.

8. Pg. 23338, Line 24: replace 'Data was...' with 'Data were...'. Corrected. 9. Pg. 23339, Lines 1-2: What are the average truncation corrections for wet and dry season aerosols? These particles would be expected to be of very different sizes and thus the magnitude of the truncation corrections should be different.

The average truncation corrections for wet and dry season were similar:  $1.15 \pm 0.09$  and  $1.10 \pm 0.06$ , at 550 nm. This is because the truncation error correction proposed by Anderson & Ogren (1998) is a linear function of the Ångström exponent:  $e = 1.337 - (0.138 \times \text{Ångström})$ . If one considers, for example, two Ångström exponent values of 1.0 and 2.0, the resulting truncation error would differ only by 13%. The average Ångström exponents for wet and dry season were compatible within one standard deviation (1.48 ± 1.12 and 1.7 1± 0.41, respectively, as shown on Table 6), and that is the reason why the average truncation corrections did not show a discernible seasonal variation.

10. Pg. 23340, Lines 2-4: Was a comparison of the T and P sensors used in this study performed so that the conversion to STP conditions during the period the nephelometer was broken was consistent with the conversion during the other study periods? The temperature inside the nephelometer is noticeably warmer than the temperature at the nephelometer inlet due to heating of the scattering volume by the lamp. Is this T appropriate for adjusting the absorption measurement to STP?

No, unfortunately a comparison between the temperature and pressure sensors was not made. However, the average STP correction factors were statistically compatible throughout the sampling period (Aug-Dec):  $1.134 \pm 0.011$  (2008 Aug-Dec, when the STP factor was based on ambient pressure and aerosol flow temperature from the diffusion dryer);  $1.144 \pm 0.011$  (2009 Aug-Dec, when STP factor was based on Nephelometer internal temperature and pressure);  $1.148 \pm 0.005$  (2010 Aug-Dec, when STP factor was based on Nephelometer internal temperature and pressure). The second part of the question was already addressed in a previous comment of the referee #1: as mentioned on line21, page 23339, the MAAP operated in series with the Nephelometer. Based on the sample flows (16.7 lpm) and on the dimensions of the tube connecting the instruments (0.432 ID and 15 cm), we estimate a sample traveling time in the order of 0.01 s. We consider that this traveling time is not enough for a significant temperature decrease in the way between the Nephelometer and the MAAP, and, therefore, we sustain that the temperature measured inside the Nephelometer is the best estimative for the sample temperature inside MAAP.

11. Pg. 23341, Lines 25-28: Diesel generator emissions...

We believe that this comment refers to the issue about the use of the term "pristine". This was addressed in a previous comment.

12. Pg. 23344, Lines 1-2: This reference is for remotely sensed aerosol parameters (i.e., AERONET-derived) which are of relatively high uncertainty and only valid for larger optical depths. Should make reference to typical values as determined by in situ studies.

We agree, and rephrased as follows: "In situ observations of dry aerosol SSA typically show values in the range 0.80-0.98 for urban aerosols (Anderson et al., 2003), 0.72-0.88 for fresh biomass burning smoke (Magi et al., 2003) and 0.88-0.99 for coarse mode dominated mineral dust (Anderson et al., 2003)."

13. Pg. 23344, Line 5: One of the earliest references for aerosol forcing efficiency is: Sheridan, P.J. and Ogren, J.A., J. Geophys. Res., 104, D14, 16793-16805, 1999.

We appreciated the reference indication, and included it in the manuscript.

- 14. *Pg. 23346, Line 7: What is the imaginary part of the refractive index? Is it 0i?* Yes, the imaginary part is 0i, and we added this information to the referred sentence.
- 15. Pg. 23346, Lines 10-28: This seems like a fairly complicated correction scheme. Do you have any references (i.e., previously published works) for this? Readers may wish to know the details of how this is done.

Iteration schemes similar to the one we used for refractive index calculation have been used before. We added the following sentence in the end of section 3.2: "Similar iterative methods for aerosol refractive index retrieval have been reported before, e.g., Guyon et al., 2003; Hand and Kreidenweis, 2002; Mack et al., 2010."

- 16. Pg. 23347, Line 15: '... 30 to 3 h.' I assume the authors mean 30 min to 3 h. Indeed, that is what we meant. We corrected, accordingly.
- 17. Pg. 23347, Lines 16-18: How were the data scrutinized? Was there a threshold for particle number, above which the data were considered contaminated by local pollution? If so, what was that threshold? How can you ensure that you are not eliminating valid data from other aerosol sources (e.g., biomass burning events) that originate from the same wind direction?

The criteria used to detect the contamination of local pollution were: i) wind blowing from 270-340°; ii) a consistent enhancement on aerosol number concentration (increase rate > 500 cm<sup>-3</sup> h<sup>-1</sup>) and/or on absorption coefficient (increase rate > 0.2 Mm<sup>-1</sup> h<sup>-1</sup>) and iii) increase and decrease within a time scale ranging from 30 minutes to 3 hours. The distinction between local pollution and biomass burning events can be made through the time scale of the perturbations in the time series. The transport of the Manaus urban plume has a spatial scale of 100 km, and its effect at the measurement site typically lasted from 4 hours to a whole day. On its turn, the regional transport of biomass burning has a spatial scale of hundreds of kilometers, and thus its temporal scale is larger, lasting from days to months.

We believe that these criteria reduce the probability of valid data elimination, although we cannot ensure 100% that no valid data was removed by mistake. In any case, the removed data corresponded to only 2% of the whole dataset, a small share to generate bias in the results. Moreover, we reasoned that this is better to be conservative and over-remove contaminated data than to have the results biased by local pollution effects, most of them a consequence of the field experiment itself.

To elucidate the criteria we used for data removal, we rephrased the referred sentence as follows: "Local pollution episodes were characterized by abrupt changes in particle number concentrations (increase rate > 500 cm<sup>-3</sup> h<sup>-1</sup>) and absorption coefficients (increase rate > 0.2 Mm<sup>-1</sup> h<sup>-1</sup>), typically lasting from 30 min to 3 hours.".

18. Pg. 23348, Lines 25-27: Is the elemental composition of African soil different than that of Amazonian soil? How can you be sure that the '... increased concentrations of crustal elements Al, Si, Ti and Fe on fine mode aerosol filter samples.' Is due to African mineral dust advection?

In the Amazon Basin, soil dust particles are not expected to be produced in significant amounts below the canopy, since the in-canopy wind velocity is low and the forest soil is covered by dead leaves, efficiently suppressing the soil re-suspension. Several evidences support the long range transport of African mineral dust to the Amazon Basin. A major transport route of wind-blown mineral dust from Africa is across the North Atlantic Ocean westward toward the American continent between February and May, when the ITCZ crosses the central part of the Amazon basin (e.g., Prospero et al., 1981; Swap et al., 1992). Early in the 1990's, ground-based measurements in the Amazon Basin indicated the occurrence of large concentrations pulses of alkali and alkaline earth elements in aerosol samples (Artaxo et al., 1994; Talbot et al., 1990). Amazonian soils are highly weathered and exhibit deficits of these elements, suggesting a non-local source for the dust (Swap et al., 1992). In the following decade, other studies based on in situ observations and remote sensing supported the contribution of African advection to the aerosol mass in Northern and Central Amazonia (e.g. Formenti et al., 2001; Kaufman et al., 2005; Koren et al., 2006). More recently, a case study reported by Ben-Ami et al. (2010) describes in detail the long range transport of African mineral dust, from the emission over West Africa, the crossing of the Atlantic Ocean, to the observed effects above the Amazon canopy about 10 days after the emission. The authors report that the arrival of the dusty air parcel over the Amazon forest increased the average concentration of aerosol crustal elements by an order of magnitude. The study combined space-borne and ground data with reanalysis model data and surface measurements, resulting in a convincing proof of the transport of African dust to the Western Amazonia. As mentioned on Page 23348, lines 22-25, other recent studies give further confirmation of the phenomenon (Ansmann et al., 2009; Baars et al., 2011), and all those references support the hypothesis that increased concentrations of crustal elements in the fine mode are associated with African dust advection.

19. Pg. 23349, Lines 14-22: Why did the median scattering ratio (2009/2010) decrease more than the median absorption ratio (2009/2010), if there were less fires in 2010 than in 2009? Wouldn't you expect a greater decrease from 2009 to 2010 in light absorption if there were fewer fires in 2010, assuming black carbon is a combustion aerosol from these fires?

This is not true that there were fewer fires in 2010 compared to 2009, as showed on Table 3. In the dry season of 2009 there was an increase in the number of fire spots in

districts neighbor (30-60 km away) and eastern to the forest reserve. In the dry season of 2010 there was a major drought, resulting in enhanced number of fire spots in the entire Brazilian Amazon, but not nearby the measurement site. Thus, what distinguishes the dry seasons of 2009 and 2010 is the proximity of fire spots and age of smoke plume. We recognize that this issue should be further discussed in the manuscript, and added a couple of sentences, as stated on comment #10 from referee #2.

20. Pg 23350, Lines 9-11: 'Increased mass fractions of these elements between January and March 2010 suggest the advection of mineral dust and biomass burning aerosols from Africa.' This is speculation. These elements could be from local soil sources or from certain urban emissions (e.g., power plants). Unless you know that the elemental signature of African soils is significantly different from that of the local soils, you can't be sure of the origin of the soil dust.

Please refere to comment #18.

21. Pg. 23351, Lines 20-22: How were the AERONET data adjusted to 637 nm wavelength?

We recognize that this information is missing, and rephrased the sentence as follows: "Considering 129 data points in northern Amazonia, the average AERONET SSA at 637 nm, <u>interpolated through a power law relationship between 441 and 673 nm</u>, is 0.91 ± 0.03 ...".

22. Pg. 23352, Lines 5-13: The suggestion that photochemical formation of secondary organic aerosols is the primary particle formation mechanism during the wet season requires one to believe that this occurs to a great extent during periods of extensive cloud cover (i.e., low light conditions). Do the kinetics of photochemical particle formation support this suggestion?

Please refer to the answer to the comment #13 by referee #1.

- 23. Pg. 23353, Line 6: replace 'estimative' with 'estimation'. Corrected.
- 24. Pg. 23353-23355, Section 4.3: I'm not sure it makes sense to compare the size distribution data, which are limited to particles below 500 nm mobility diam, with the optical properties (angstrom exponent and backscattering fraction), which include contributions from much larger (PM7) particles. How would the relationships you have determined be affected by the mismatched size ranges used for comparison?

Please refer to comment #15 by referee #1.

25. Figures: All of the figures should be remade at higher resolution. These were difficult to see clearly and are not of publication quality.

All figures were improved regarding to resolution. Some of them were also reformulated, as follows.

### **Updated figures**

Figure 1: The yellow circle marks the location of the TT34 measurement tower in the Sate of Amazonas, Brazil. The big red circles mark the position of some of the major cities in the Brazilian North and Northeast regions (more than 1.4 million inhabitants each). The small red circles mark the position of municipalities neighbor to the forest reserve (Rio Preto da Eva, Presidente Figueiredo, Novo Airão), and municipalities eastern to the forest reserve in the State of Amazonas (Barreirinha, Itapiranga, Nhamundá, Parintins, São Sebastião do Uatumã, Silves and Urucará).



Figure 2: Monthly accumulated precipitation measured at INPA's K34 tower from January 2008 to June 2011. The line represents the percent data coverage for each month.



Figure 3: a) Wind rose plots for the period between January 2008 and June2011. b) Median aerosol particle number concentration as a function of wind direction for the wet season (Jan-

Jun) and for the dry season (Jul-Dec). The Manaus city is located 60 km away in the southeast direction.



Figure 4: Daily medians of aerosol particle scattering coefficients at 550 nm (a) and of aerosol particle absorption coefficients at 637 nm (b), from Feb 2008 to May 2011. Error bars represent first and third quartiles. Shaded areas represent the dry season period.





Figure 5: Elemental concentrations of fine mode (PM2) crustal elements (AI, Si, Ti, Fe) and potassium, and particle absorption coefficients at 637 nm averaged according to elemental composition integration times. Only data between January and May of each year is shown.

Figure 6: Daily averages of aerosol optical depth (AOD) observations from MODIS (TERRA-AQUA), AERONET (Manaus) and in situ extinction measurements above the canopy. AOD observations from MODIS were integrated inside an area with 40 km radius around the in situ measurement site. AOD observations from AERONET are level 2.0 in 2008 Jul-Oct and level 1.5 in 2011 Jan-Apr, and were interpolated to 550 nm using AOD(500) and the Ångström exponent between 440 and 675 nm. Shaded areas represent the dry season period.



Figure 7: Median 2008-2011 seasonal cycle for particle scattering coefficients at 550 nm, particle absorption coefficients at 637 nm, and particle single scattering albedo (SSA) at 637 nm. Statistics were calculated for each 10 Julian days. The box lines represent the lower quartile, median and upper quartile values. Whiskers extend within 1.5 times the interquartile range.



Figure 8: Monthly averages of AERONET retrieved single scattering albedo between 1993 and 2011 at seven different locations in Amazonia: Ji Paraná, Alta Floresta, Rio Branco (arc of deforestation), Balbina, Belterra, Santarém, Manaus (northern Amazonia). All data is level 2, with exception to 9 Manaus data points in 2008, which are level 1.5. Particle SSA values observed in the AERONET wavelengths were interpolated to 637 nm. Error bars indicate standard deviations.



Figure 9: Local time diurnal cycle boxplots for wet season (a) aerosol particle scattering; (b) aerosol particle absorption. The box lines represent the lower quartile, median, and upper quartile values.



Figure 10: Time series of particle mass scattering coefficient at 550 nm, Particle mass absorption coefficient at 637 nm, and ratio between PM2 and PM10 particle mass, observed from gravimetric analysis of 199 stacked filter units samples. Shaded areas represent the dry season period.



Figure 11: Relationships between scattering Ångström exponents and three parameters calculated from sub micrometer aerosol particle number size distributions (10-500 nm): count mean diameter (CMD), surface area mean diameter (SMD) and volume mean diameter (VMD). The plots comprise measurements taken between July and August 2009.



Figure 12: Frequency histograms for the aerosol forcing efficiency and for the cloud fraction during wet and dry seasons, comprising the period between November 2009 and September 2010.



Figure 13: Cumulative contribution of 13 particle size ranges to scattering (550 nm) and absorption (637 nm) coefficients calculated using a Mie model. Error bars represent standard deviations.



Figure 14: Example of Manaus urban plume entrance in the research site on 2009 April 25<sup>th</sup>. a) Evolution of particle single scattering albedo – SSA (637nm), scattering coefficient (550 nm), absorption coefficient (637 nm) and number concentration; b) HYSPLIT back-trajectories reaching the measurement site at the time specified (local time). Each trajectory point represents one hour less from the start time.



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