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# Downwelling solar irradiance in the biomass burning region of the southern Amazon: Dependence on aerosol intensive optical properties and role of water vapor

Nilton E. Rosário,<sup>1,2</sup> Marcia A. Yamasoe,<sup>1</sup> Helen Brindley,<sup>2</sup> Thomas F. Eck,<sup>3,4</sup> and Joel Schafer<sup>3</sup>

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[1] The sensitivity of solar irradiance at the surface to the variability of aerosol intensive optical properties is investigated for a site (Alta Floresta) in the southern portion of the Amazon basin using detailed comparisons between measured and modeled irradiances. Apart from aerosol intensive optical properties, specifically single scattering albedo ( $\omega_{o\lambda}$ ) and asymmetry parameter ( $g_\lambda$ ), which were assumed constant, all other relevant input to the model were prescribed based on observation. For clean conditions, the differences between observed and modeled irradiances were consistent with instrumental uncertainty. For polluted conditions, the agreement was significantly worse, with a root mean square difference three times larger ( $23.5 \text{ Wm}^{-2}$ ). Analysis revealed a noteworthy correlation between the irradiance differences (observed minus modeled) and the column water vapor (CWV) for polluted conditions. Positive differences occurred mostly in wet conditions, while the differences became more negative as the atmosphere dried. To explore the hypothesis that the irradiance differences might be linked to the modulation of  $\omega_{o\lambda}$  and  $g_\lambda$  by humidity, AERONET retrievals of aerosol properties and CWV over the same site were analyzed. The results highlight the potential role of humidity in modifying  $\omega_{o\lambda}$  and  $g_\lambda$  and suggest that to explain the relationship seen between irradiances differences via aerosols properties the focus has to be on humidity-dependent processes that affect particles chemical composition. Undoubtedly, there is a need to better understand the role of humidity in modifying the properties of smoke aerosols in the southern portion of the Amazon basin.

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## 1. Introduction

[2] The downwelling solar irradiance at the surface is a crucial component of atmospheric circulation models [Wild, 1999, 2005; Zamora *et al.*, 2005]. Recently, the use of crude parameterizations of aerosol optical properties has emerged as a major source of discrepancy in the modeling of downwelling solar irradiance at the surface, in particular in areas characterized by high aerosol loadings [Wild, 1999, 2005]. The relative contributions of the absorption and scattering processes to the aerosol extinction and the geometry of scattering, usually expressed in terms of single

scattering albedo ( $\omega_{o\lambda}$ ) and asymmetry parameter ( $g_\lambda$ ), are critical to the modeling of the impact of aerosols on the downwelling solar irradiance at the surface. Explicit modeling of  $\omega_{o\lambda}$  and  $g_\lambda$  is still a difficult and computationally costly task [Chin *et al.*, 2009] due to their dependence on complex and dynamic processes, such as emission, aging, mixture state, water vapor uptake, cloud processing etc [Menon, 2004; Reid *et al.*, 2005]. Consequently, very often aerosol properties are fixed in space and time in atmospheric circulation models based on typical values. This approach can be critical in certain scenarios, especially those where aerosol intensive properties are highly variable, such as biomass burning areas [Wild, 1999, 2005; Reid *et al.*, 2005].

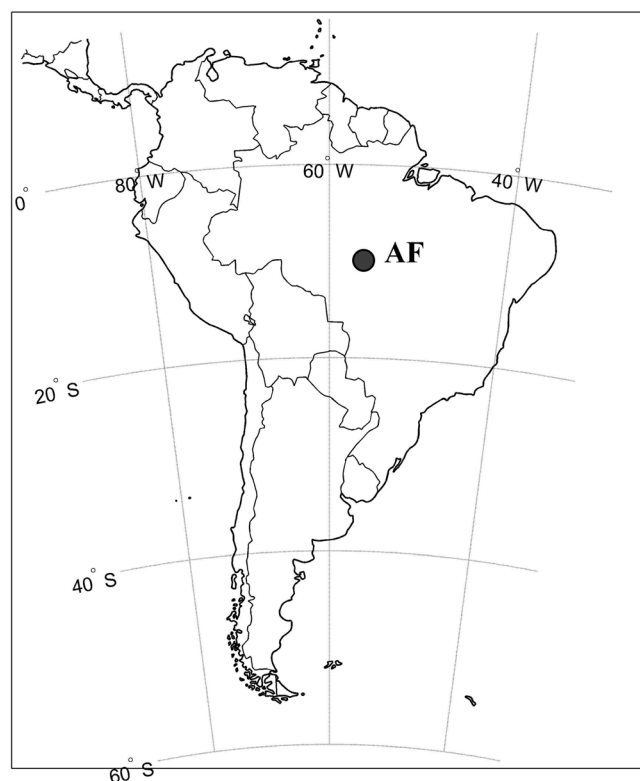
[3] In the present paper, we use differences between model-calculated and measured downwelling broadband solar irradiances at the surface for a site in the main Amazon's biomass burning area to analyze the impacts of neglecting the variability in  $\omega_{o\lambda}$  and  $g_\lambda$ . The irradiance measurements were taken from a SolRad-Net (Solar Radiation Network [Schafer *et al.*, 2002]) pyranometer operated alongside an AERONET (Aerosol Robotic Network

<sup>1</sup>Atmospheric Sciences Department, University of São Paulo, São Paulo, Brazil.

<sup>2</sup>Physics Department, Imperial College London, London, UK.

<sup>3</sup>Biospheric Sciences Branch, NASA GSFC, Greenbelt, Maryland, USA.

<sup>4</sup>GEST Center, University of Maryland Baltimore County, Catonsville, Maryland, USA.



**Figure 1.** Geographical localization of the Alta Floresta site.

[Holben *et al.*, 1998]) Sun photometer installed in the southern portion of the Amazon basin. Section 2 provides a brief description of the experimental setup, the data set and the methodology. Section 3 presents the results, which consist of an evaluation of the agreement between measurements and model-calculations for aerosol background conditions, for which one would expect that the impact of aerosol optical properties variability would be marginal, and for polluted conditions. For the latter case, the performance of the radiative transfer (RT) model was analyzed in detail, focusing on the impact of the variability of  $\omega_{o\lambda}$  and  $g_{\lambda}$ , and on the dependence of the impact on aerosol loading and atmospheric humidity. Moreover, AERONET long-term retrievals of aerosol intensive optical and microphysical properties over Alta Floresta were analyzed. Final considerations are presented in section 4.

## 2. Experimental Setup

### 2.1. Experimental Site

[4] The Alta Floresta site (Lat. 09° 52'S; Lon. 56° 06'W; Elev. 277 m, Figure 1) is located in the southern region of the Amazon basin in a transition zone from cerrado to primary forest, surrounded by areas heavily impacted by biomass burning. During the dry season and before the biomass burning season, specifically from April to June, biogenic emissions and soil dust resuspension are the main local sources of aerosol in the southern portion of the Amazon basin [Echalar *et al.*, 1998; Maenhaut *et al.*, 2002], and the average aerosol optical depth (AOD) in the visible ( $\lambda = 550$  nm) is approximately 0.10 [Procopio *et al.*, 2004]. After

the beginning of the burning season, emissions from biomass burning become the main source of aerosols to the regional atmosphere. The average AOD during the burning season has a significant inter-annual variability, in 2001 the mean AOD at 500 nm over Alta Floresta was 0.61 and in 2002 the value increased to 1.14 [Procopio *et al.*, 2004]. Instantaneous values of aerosol optical depth in the visible spectral region often reach values as high as 3.0 [Schafer *et al.*, 2008; Rosario *et al.*, 2009].

### 2.2. Data Set

[5] The main observational data set consists of 1662 cloud-free coincident measurements taken in a broad range of AOD conditions by a pyranometer (Kipp & Zonen CM 21) and a Sun-tracking photometer (Cimel Eletronique CE318) in direct Sun mode during the dry and dry-to-wet seasons (from May to November) of the years 2000, 2001 and 2002. The pyranometer is part of the SolRad-Net (Solar Radiation Network) [Schafer *et al.*, 2002] and the photometer belongs to the Aerosol Robotic Network (AERONET) [Holben *et al.*, 1998]. The measurements were taken from the Level 2.0 databases of both networks, which are quality assured. The absolute uncertainty of the pyranometer is stated as 2% and the estimated accuracies of spectral aerosol optical depth (440, 670, 870, and 1020 nm) and column water vapor (CWV) provided by the Sun/sky photometer are 0.01–0.02 [Eck *et al.*, 1999] and ~5–10% [Halthore *et al.*, 1997; Smirnov *et al.*, 2004], respectively. Collocated retrievals of column ozone from the Total Ozone Mapping Spectrometer (TOMS; [http://toms.gsfc.nasa.gov/ozone/ozone\\_v8.html](http://toms.gsfc.nasa.gov/ozone/ozone_v8.html)) and broadband surface albedo ( $\alpha_{sfc} = 0.13$ ) values taken from the study of Berbet and Costa [2003] for the Amazon basin were also used as input parameters to the RT model. Sensitivity analyses showed that the difference between model calculated solar broadband irradiance at the surface using broadband or spectrally varying albedo is lower than instrumental uncertainty. Climatologies of  $\omega_{o\lambda}$  and  $g_{\lambda}$  were calculated from AERONET inversion products version 2 and level 2, which use the algorithm and input albedo data described respectively by Dubovik and King [2000] (as updated by Dubovik *et al.* [2006]) and described by Eck *et al.* [2008], for the period of the present study, 2000–2002, and for much longer period between 1999 and 2007.

### 2.3. Radiative Transfer Model

[6] The Santa Barbara DISORT Atmospheric Radiative Transfer code (SBDART) [Ricchiazzi *et al.*, 1998] was used to simulate the downward solar irradiance at the surface. The performance of SBDART has been evaluated against other radiative transfer models and high quality measurements. Regarding downward broadband solar irradiance at the surface, its consistency with other models and high quality measurements has been shown to be better than 2% [Halthore *et al.*, 2005; Michalsky *et al.*, 2006]. It is worth mentioning that most of these evaluations were performed for either clean or moderate pollution conditions when compared with the highly polluted conditions seen over the Amazon basin during the biomass burning season. However, Procopio *et al.* [2003] compared measured and modeled downward spectral radiance at the surface in the Amazon basin using SBDART for heavy smoke conditions and found good agreement. The code offers different options

**Table 1.** Mean, Standard Deviation (std), Maximum (max) and Minimum (min) of Single Scattering Albedo ( $\omega_{o\lambda}$ ) and Asymmetry Parameter ( $g_\lambda$ ) From AERONET Sky Inversion Retrievals Over Alta Floresta for Long-Term (1999–2007) and for the Period Analyzed in the Current Study (2000–2002)

Variable	Statistic	440 nm	670 nm	870 nm	1020 nm
<i>1999–2007</i>					
$\omega_{o\lambda}$	mean	0.93	0.92	0.90	0.88
	std	0.02	0.04	0.05	0.05
	max	0.99	0.99	0.99	0.99
	min	0.83	0.80	0.72	0.64
$g_\lambda$	mean	0.68	0.57	0.52	0.49
	std	0.02	0.03	0.04	0.05
	max	0.75	0.68	0.65	0.67
	min	0.61	0.49	0.42	0.39
<i>2000–2002</i>					
$\omega_{o\lambda}$	mean	0.93	0.92	0.91	0.90
	std	0.02	0.03	0.04	0.05
	max	0.99	0.99	0.99	0.98
	min	0.83	0.80	0.79	0.78
$g_\lambda$	mean	0.68	0.58	0.52	0.49
	std	0.02	0.03	0.04	0.05
	max	0.75	0.68	0.65	0.65
	min	0.62	0.51	0.43	0.39

to prescribe aerosol radiative properties. In the present study spectral aerosol optical depth, from AERONET instantaneous observations, single scattering albedo and asymmetry parameter, at AERONET standard wavelengths (440, 670, 870 and 1020 nm) were used as input data to run the code. For further details about SBDART aerosol prescription refer to Ricchiuzzi *et al.* [1998].

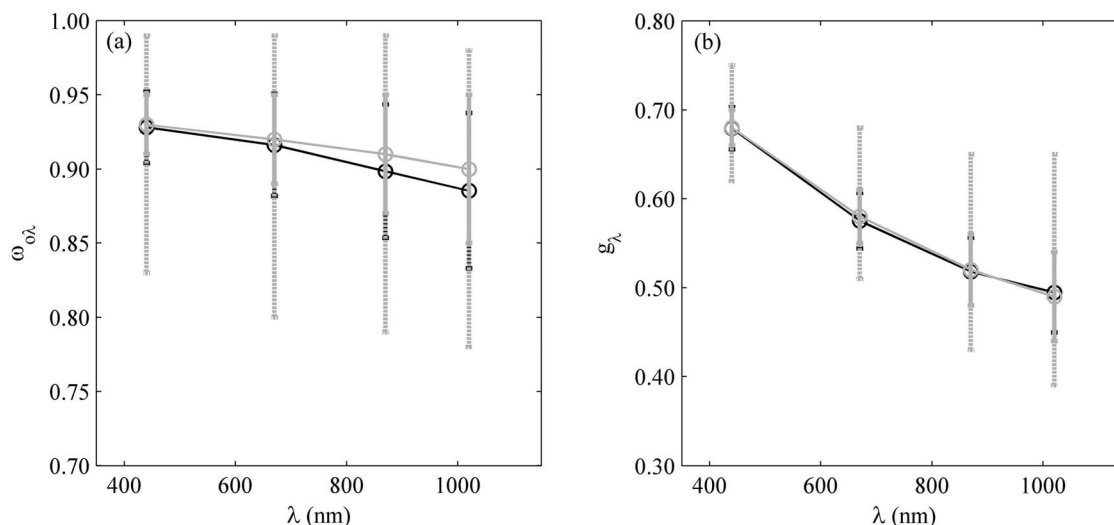
## 2.4. Cloud Screening Procedures

[7] The AERONET direct Sun measurements' cloud screening procedure [Smirnov *et al.*, 2000] is implicitly employed by choosing solar irradiance measurements that are coincident with the Level 2 Sun photometer direct Sun retrievals. However, although this “assures” absence of cloud along the path of the Sun direct beam, the selection of simultaneous measurements does not avoid a possible cloud effect on the diffuse component. According to Gu *et al.* [2001] the cloud enhancement effect on the diffuse solar component can increase ground surface irradiance by more than 20% and last for periods of up to 30 min. As an attempt to avoid this effect, the variability of the measured irradiance at the surface, as expressed by the standard deviation for an interval of 8 min centered on the AERONET retrieval, was stipulated not to exceed the pyranometer uncertainty (2%). This filter was expected to reduce cloudy cases by avoiding large temporal variance in the solar irradiance at the surface. Roughly 25% of coincident pyranometer–Sun photometer measurements met this criterion. This is not surprising as the probability of observation of cloudless skies over vast areas in the Amazon basin is very low. Analyzing cloud cover using Landsat TM scenes, Asner [2001] showed that even during the dry season the probability of occurrence of cloud fraction below 20% in a Landsat footprint in the southern region of the Amazon basin is less than 40%. As a further test, the remaining measurements of downwelling solar irradiance at the surface

were analyzed against model-calculated results assuming a purely scattering aerosol ( $\omega_{o\lambda} = 1.0$ ) and considering the maximum  $g_\lambda$  retrieved by AERONET over Alta Floresta (Table 1), a scenario in which one would expect to see the maximum downwelling solar irradiance at the surface for a given aerosol optical depth. Irradiance measurements exceeding the maximum theoretical downwelling irradiance at the surface by more than the pyranometer uncertainty were excluded. This criterion excluded less than 10% of the previous remaining cases.

## 2.5. Methodology

[8] This study was primarily based on the analysis of the differences observed between measured and model-calculated broadband downwelling solar irradiance at the surface in cloudless skies. The aim was to investigate how the variability in aerosol intensive optical properties modulate the downward solar irradiances by keeping  $\omega_{o\lambda}$  and  $g_\lambda$  fixed while specifying all other relevant model input parameters such as spectral aerosol optical depth, water vapor amount and ozone content using coincident observations. The  $\omega_{o\lambda}$  and  $g_\lambda$  were fixed using the average of instantaneous retrievals of these quantities from the AERONET Sun-tracking photometer almucantar retrievals over the same period, which produced similar results compared with averages taken over a much longer period, i.e., from 1999 to 2007 (Figure 2). Assuming that the RT model is capable of capturing the effects of all other relevant parameters accurately, one would expect that when the aerosol loading increases, statistically significant discrepancies between measured and model-calculated solar irradiance at the surface would be dominated by the variability in  $\omega_{o\lambda}$  and  $g_\lambda$ . Figure 3 shows the results of a sensitivity test of downwelling solar irradiance at the surface to the variability of  $\omega_{o\lambda}$  and  $g_\lambda$  over Alta Floresta according to the values shown in Table 1. The results are presented in terms of relative difference between the model-calculated downwelling solar irradiances at the surface using varying and mean intensive optical properties. Figure 3a shows differences between modeled downwelling solar irradiance at the surface using mean values and modeled downwelling solar irradiance at the surface using maximum and minimum values of  $\omega_{o\lambda}$  and  $g_\lambda$  as a function of solar zenith angle. Figure 3b presents the same evaluation assuming standard deviations as a metric of variability. It is shown that for  $AOD_{440\text{ nm}} \leq 0.1$  (background condition), even considering the maximum and minimum observed values of  $\omega_{o\lambda}$  and  $g_\lambda$ , the impact on downwelling solar irradiance at the surface does not exceed the pyranometer accuracy. Therefore, measurements obtained under these conditions were used to evaluate the agreement between observed and model-calculated irradiances when the impact of the variability of aerosol properties is negligible. The remaining measurements ( $AOD_{440\text{ nm}} > 0.1$ ) were divided in two groups according to AOD at 440 nm. The upper threshold ( $AOD_{440\text{ nm}} \geq 0.4$ ) was chosen based on the standard AERONET threshold which aims to assure the accurate retrieval of single scattering albedo [Dubovik and King, 2000; Dubovik *et al.*, 2002]. At the peak of the biomass burning season (from August to September) events of  $AOD_{440\text{ nm}} \geq 0.4$  over Alta Floresta represent roughly 30% of Sun-tracking photometer retrievals in direct Sun



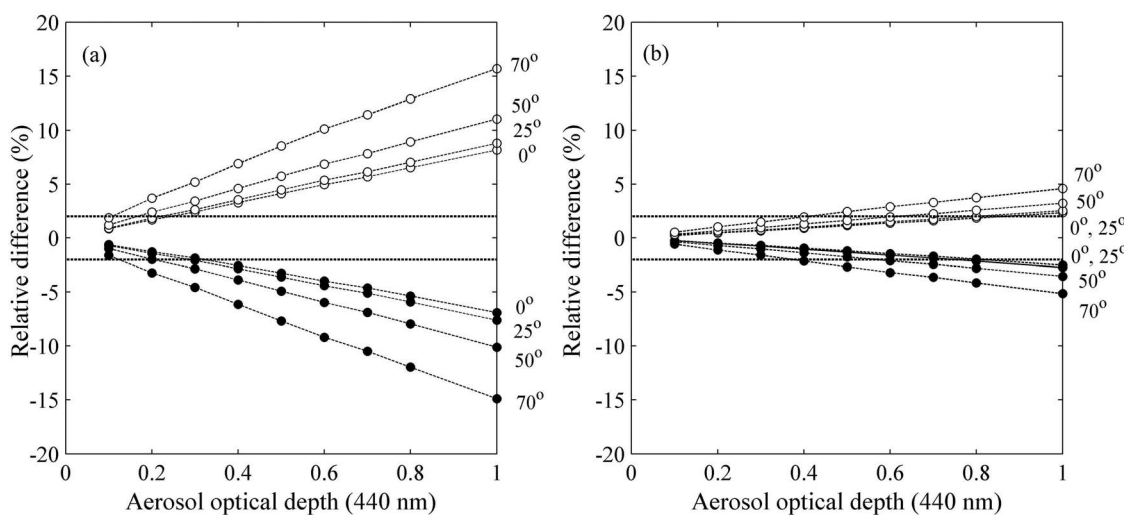
**Figure 2.** Spectral dependency of (a) single scattering albedo and (b) asymmetry parameter as obtained from average and standard deviations of AERONET retrievals over Alta Floresta for the period covered by the present study (2000–2002, gray) and for long-term retrievals (1999–2007, black). The gray dashed lines represent the maximum and minimum values according to the retrievals for the analyzed period.

mode. The variability of instantaneous  $\omega_{o\lambda}$  and  $g_{\lambda}$  was also analyzed in order to evaluate the consistency of our hypothesis, that variations in aerosol intensive properties might explain most of the differences between measured and model-calculated irradiance. According to Reid *et al.* [2005], variability in  $\omega_{o\lambda}$  and  $g_{\lambda}$  in the southern portion of the Amazon Basin during the biomass burning season are essentially controlled by variability in the fine mode microphysical parameters. A Mie code was used to assess the sensitivity of  $\omega_{o\lambda}$  and  $g_{\lambda}$  to the observed variability of fine mode volume median radius ( $r_{fm}$ ), fine mode geometric

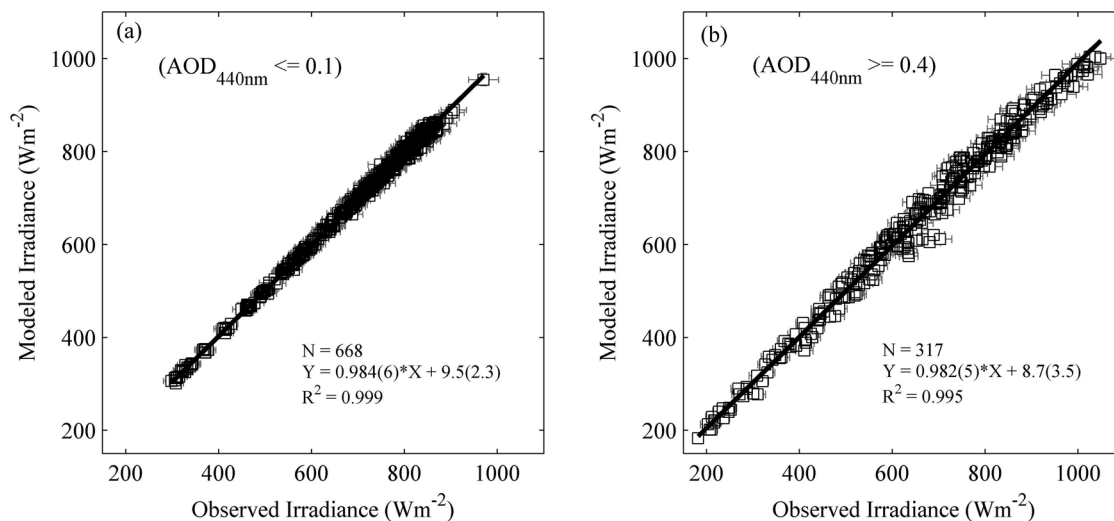
standard deviation ( $\sigma_{fm}$ ) and complex refractive index ( $n + ik$ ) over Alta Floresta.

### 3. Results

[9] Figure 4 shows scatterplots comparing measured and model-calculated downwelling solar irradiance at the surface for background ( $AOD_{440 \text{ nm}} \leq 0.1$ ) and polluted conditions ( $AOD_{440 \text{ nm}} \geq 0.4$ ). For background conditions, the differences between measured and model-calculated irradiance was within the estimated instrumental uncertainty with the differences confined to values lower than 2%, which is



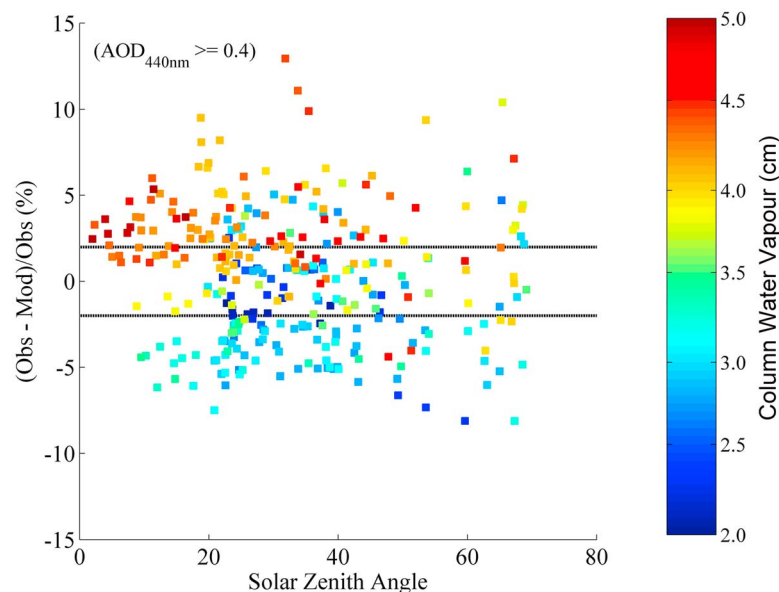
**Figure 3.** Relative difference between model-calculated solar irradiance at the surface using average values of single scattering albedo ( $\omega_{o\lambda}$ ) and asymmetry parameter ( $g_{\lambda}$ ) and model-calculated solar irradiance considering (a) maximum and minimum values of  $\omega_{o\lambda}$  and  $g_{\lambda}$  values and (b) the average values plus and minus one standard deviation. The differences are presented as function of aerosol optical depth at 440 nm and solar zenith angle.



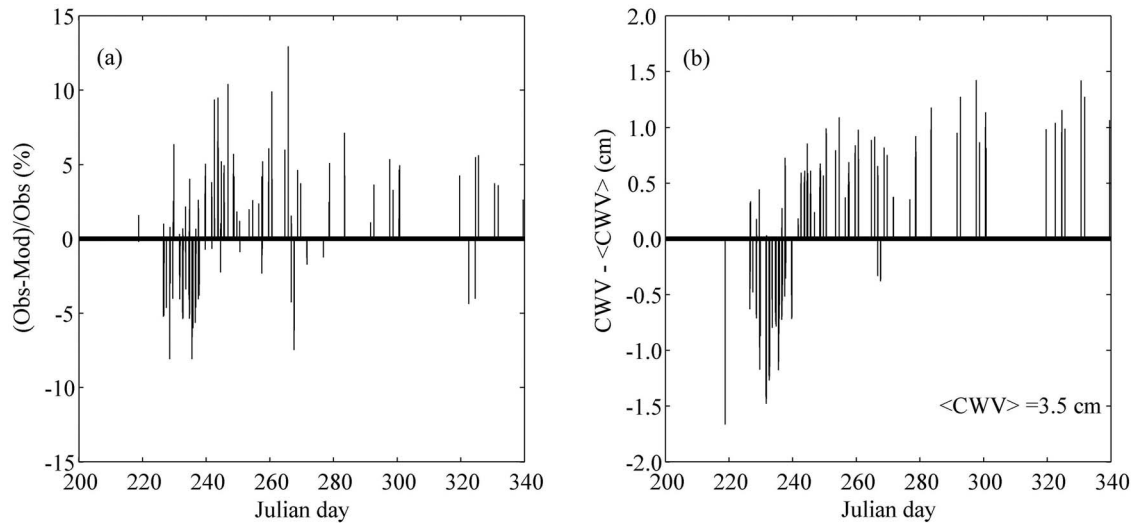
**Figure 4.** Scatterplot of measured broadband downwelling solar irradiance at the surface against model-calculated results for aerosol loadings typical of (a) background ( $AOD_{440\text{ nm}} \leq 0.1$ ) and (b) polluted conditions ( $AOD_{440\text{ nm}} \geq 0.4$ ). The errors bars represent the standard deviation of observed solar irradiance for an interval of 8 min centered on the AERONET retrieval.

consistent with results of the previous sensitivity study. Therefore, in such AOD conditions, as suggested, the impact of variability in  $\omega_{o\lambda}$  and  $g_\lambda$  is lower than measurements uncertainty. The calculated Root Mean Square Difference (RMSD) and Mean Difference (MD) are  $7.8\text{ Wm}^{-2}$  and  $2.3\text{ Wm}^{-2}$ , respectively. This level of agreement is similar to that seen in previous studies in which the performance of SBDART was evaluated [Halothore *et al.*, 2005; Michalsky *et al.*, 2006]. For polluted conditions, a significant degradation in the agreement between measured and model-calculated downwelling solar irradiance was observed. The RMSD increased by a factor of three com-

pared with background conditions ( $23.5\text{ Wm}^{-2}$ ), and relative differences reach values as high as 12%. Apart from the expected and observed dependence of the differences on aerosol loading, further analysis revealed a noteworthy pattern of behavior between the irradiance differences (observed minus modeled) and CWV for polluted conditions cases. Positive differences occur preferentially in wet atmospheres while negative values occur more frequently in drier conditions (Figure 5). Such features are not seen in low aerosol conditions (not shown). Following the CWV seasonality, the differences are characterized by a seasonal pattern, with negative values occurring in general at the



**Figure 5.** Relative difference between observed (Obs) and model-calculated (Mod) broadband downwelling solar irradiance at the surface for polluted conditions ( $AOD_{440\text{ nm}} \geq 0.4$ ) as a function of column integrated water vapor and solar zenith angle.

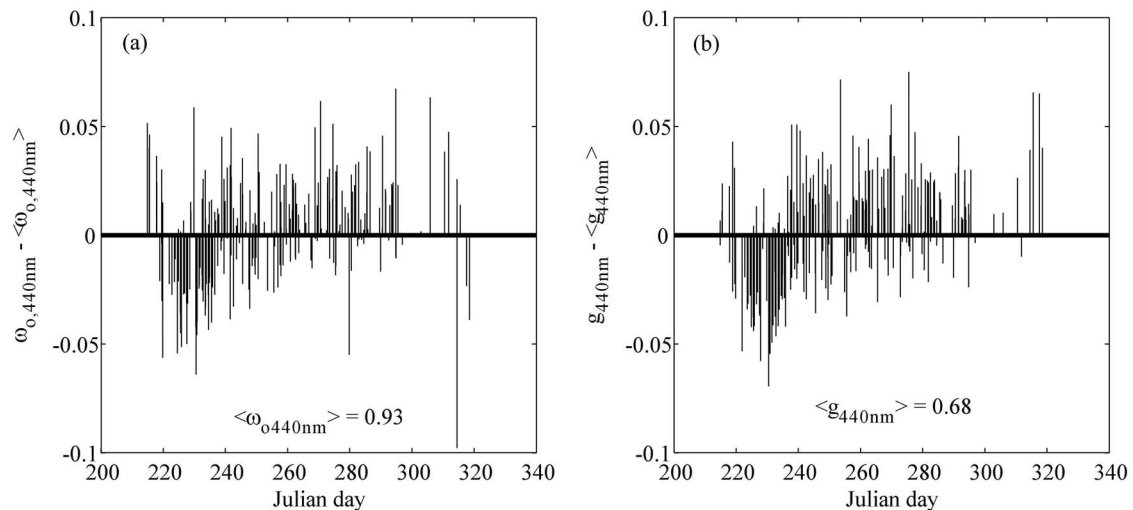


**Figure 6.** Seasonal distribution of (a) relative difference between observed (Obs) and model-calculated (Mod) broadband downwelling solar irradiance at the surface and (b) deviations of observed Column Water Vapor (CWV) from the mean value ( $\langle \text{CWV} \rangle$ ) as calculated for polluted conditions ( $\text{AOD}_{440 \text{ nm}} \geq 0.4$ ).

beginning of the burning season, when CWV is low, and positive values seen from the middle to the end, when CWV begins to increase (Figure 6). Analysis of deviations of AERONET multiyear instantaneous retrievals of  $\omega_{o\lambda}$  and  $g_{\lambda}$  from climatological values over Alta Floresta show a seasonal trend consistent with this pattern (Figure 7). Aerosols characterized by  $\omega_{o\lambda}$  and  $g_{\lambda}$  lower than climatological values are frequent at the beginning of the biomass burning season while toward the middle and the end of the burning season aerosols with larger values of  $\omega_{o\lambda}$  and  $g_{\lambda}$  become dominant. The consistency between the two independent analyses, one using pyranometer measurements and other using Sun photometer's aerosol optical properties retrievals corroborates the initial hypothesis, that once all other variables are modeled accurately, differences in irradiance would reflect the variability in aerosol optical properties. The physical dependence of downward solar irradiance at

the surface on aerosol optical properties is well established. Therefore it would appear that there is a link between the seasonality observed in the irradiance differences and the deviations of  $\omega_{o\lambda}$  and  $g_{\lambda}$  from climatological values.

[10] Nevertheless, despite their consistent seasonal evolution, a direct physical relationship between humidity and deviations of  $\omega_{o\lambda}$  and  $g_{\lambda}$ , cannot be easily established. The issue itself is complex and not well understood in the context of the Amazon basin. There are several humidity dependent processes that can affect aerosol optical properties, and they occur simultaneously with non humidity dependent processes that also influence aerosol properties. Moreover, one should take into account the likelihood of a natural concurrency or measurements artifact. Despite these challenges, we attempted to explore the nature of the seasonal correlation found between irradiance differences, humidity and aerosol optical properties. Aerosols from



**Figure 7.** Seasonal distribution of multiyear deviations (1999–2007) of (a) single scattering albedo ( $\omega_{o\lambda}$ ) and (b) asymmetry parameter ( $g_{\lambda}$ ) at 440 nm from climatological values ( $\langle \omega_{o\lambda} \rangle$ ;  $\langle g_{\lambda} \rangle$ ).

biomass burning in the Amazon basin have been suggested to have low hygroscopicity when compared with other type of aerosols [Kotchenruther and Hobbs., 1998; Martin *et al.*, 2010]. Nevertheless, as mentioned, other humidity dependent processes, such as fuel moisture content, cloud processing [Reid *et al.*, 2005] and amount of woody fuel [Eck *et al.*, 2009] are likely to affect the seasonal variability of aerosol intensive optical properties. Despite their distinct nature, it is well known that all these processes, including hygroscopicity, tend to lead to an increase in  $\omega_{o\lambda}$  and  $g_{\lambda}$  [Reid *et al.*, 2005]. At the early stage of the burning season, due to lower fuel moisture content and less amount of woody fuel burning, the percentage of biomass burning combustion that is in the flaming phase is expected to be greater than during the latter stages [Eck *et al.*, 2009], which produce smaller and more absorbing particles [Reid *et al.*, 1998]. As moisture content increase through the burning season, the percentage of combustion in the smoldering phase becomes dominant [Schäfer *et al.*, 2008], which in turn produces larger and less absorbing particles. Additionally, the increase of cloudiness during the late stages of the burning season enhances cloud processing of aerosol [Reid *et al.*, 2005]. All these processes affect particles intensive optical properties via their microphysical properties.

[11] Hence, here we analyzed AERONET retrievals of aerosol fine mode size distribution parameters, specifically volume median radius ( $r_{fm}$ ) and geometric standard deviation ( $\sigma_{fm}$ ), and complex refractive index ( $n + ik$ ). Table 2 shows the results of the numerical study using a Mie code which aimed to evaluate the sensitivity of  $\omega_{o\lambda}$  and  $g_{\lambda}$  to the extreme values of fine mode parameters and complex refractive index retrieved by AERONET over Alta Floresta. A knowledge of the main microphysical parameters that controls the variability in  $\omega_{o\lambda}$  and  $g_{\lambda}$  is an important step to characterize the potential role of the processes mentioned. The study consisted of varying a specific parameter between its extreme values while keeping the others parameters fixed according to climatological values for Alta Floresta. The results showed that the observed variability in  $r_{fm}$  alone has a low impact on  $\omega_{o\lambda}$  in the visible spectrum becoming more relevant in the near-infrared spectral region. As expected, the variability in  $r_{fm}$  controls most of the variation in  $g_{\lambda}$  in both spectral regions visible and near-infrared, whereas the variability in  $\sigma_{fm}$  only has a relevant effect on  $g_{\lambda}$  in the near-infrared region, since it is only here that the differences seen exceed the retrieval uncertainty. Of all the parameters considered, the variability in the imaginary part ( $k$ ) of the complex refractive index has the largest impact on  $\omega_{o\lambda}$  in both spectral regions. The real part ( $n$ ) of the refractive index has a larger impact on  $\omega_{o\lambda}$  in the near-infrared region when compared with the visible with the reverse observed regarding its impact on  $g_{\lambda}$ . Essentially, the Mie sensitivity study suggests that complex refractive index ( $n + ik$ ) plays a dominant role on the variability of  $\omega_{o\lambda}$  of aerosols over Alta Floresta, while  $g_{\lambda}$  variability is basically controlled by fine mode size parameters. Figure 8 shows scatterplots of  $r_{fm}$ ,  $\sigma_{fm}$ ,  $n$  and  $k$  as a function of CWV. As shown previously by Schäfer *et al.* [2008],  $r_{fm}$  tends to increase with CWV, whereas  $\sigma_{fm}$  does not present a clear trend.  $k$  has notable decreasing tendency as CWV increases and  $n$  as well. The lowering of  $n$  with CWV is the only trend that decreases  $\omega_{o\lambda}$ . However, the dominant effect of  $k$  on  $\omega_{o\lambda}$ , along with

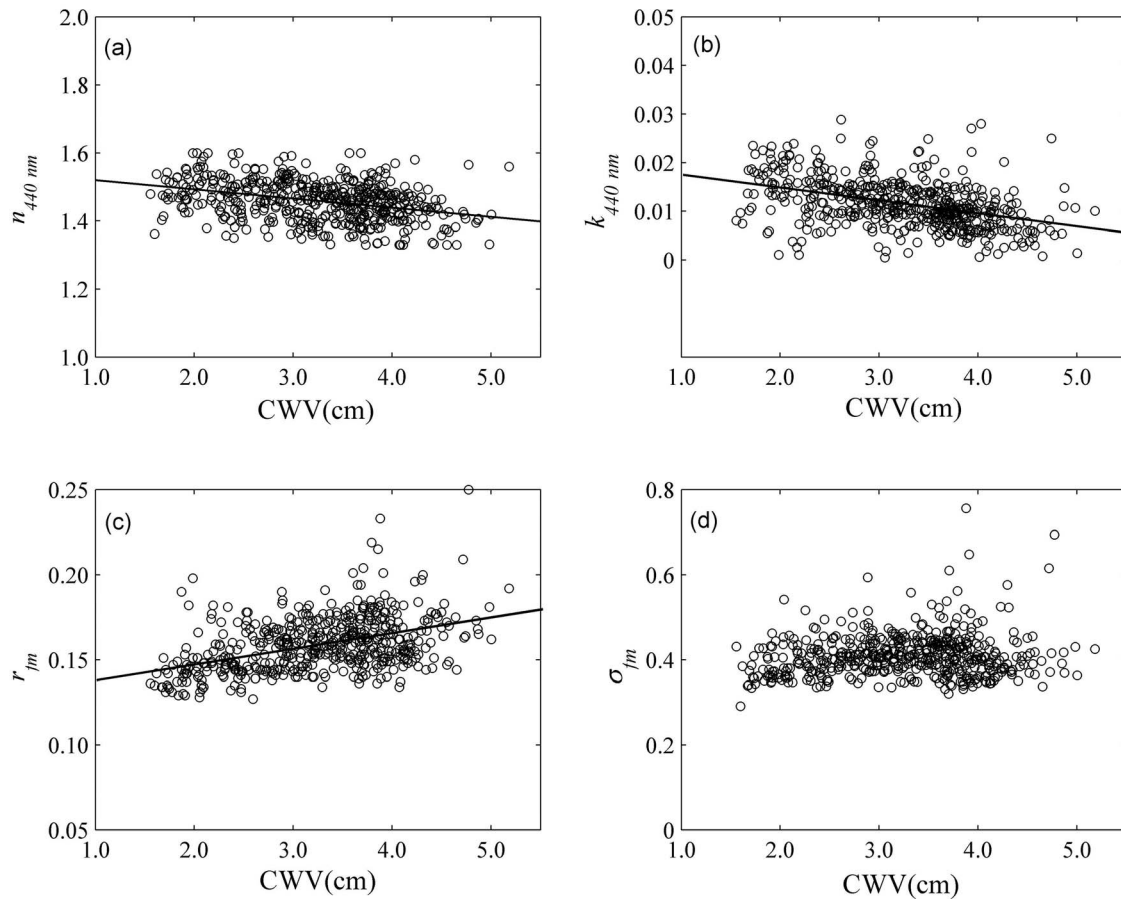
the contribution of  $r_{fm}$ , supersedes that of the variability in  $n$ . Figure 9 shows similar scatterplots of both  $r_{fm}$  and  $k$  as a function of AOD. Both microphysical parameters  $r_{fm}$  and  $k$  did not present a noticeable relation with AOD, mainly  $k$ . The weak relation between  $r_{fm}$  and AOD was somehow surprisingly, since coagulation is considered an important growth mechanism of aerosols in aged smoke plumes and it is highly depend on aerosol loading [Reid *et al.*, 1998].

[12] Unlike intensive optical properties themselves, the relationship between humidity conditions and specific microphysical parameters allows a more direct evaluation of the processes associated with optical properties variability. Here, the likely explanation for the occurrence of large  $k$  during low CWV scenarios is the low fuel moisture content, which increases combustion efficiency leading to the dominance of combustion in flaming phase, which in turn produces larger amount of black carbon, unlike smoldering combustion. The results as a whole suggest that alteration in particle chemical composition is the main controller in determining the seasonal variability in  $\omega_{o\lambda}$ , and since variations in  $\omega_{o\lambda}$  in this region have larger impact on surface irradiance than variations in  $g_{\lambda}$  [Eck *et al.*, 1998], therefore of the observed trend in irradiance differences.

#### 4. Final Remarks

[13] The impact of variability of aerosol intensive optical properties, specifically single scattering albedo and asymmetry parameter, on downwelling solar irradiance at the surface for a site located in a biomass burning region in the Amazon basin was investigated using differences between observed and model-calculated solar irradiances at the surface. Apart from single scattering albedo and asymmetry parameter, which are assumed static, all other relevant input to the modeling of the downwelling solar irradiance at the surface were accurately prescribed based on observations. The difference between observed and model-calculated solar irradiances was highly dependent on aerosol loading. For background aerosol loading ( $AOD_{440\text{ nm}} \leq 0.1$ ), differences were in general within experimental uncertainties, whereas for polluted conditions ( $AOD_{440\text{ nm}} > 0.4$ ) differences reached values well above uncertainties, and RMSD was three times higher ( $23.5\text{ Wm}^{-2}$ ) than for the background conditions. Detailed analysis revealed a noteworthy relationship between column water vapor content and the differences between observed and model-calculated solar irradiances at the surface. In wet conditions, the differences tended to be positive, while the opposite was observed in drier circumstances. Such a pattern of behavior was not observed for background loading conditions. Given this dependence on humidity, for polluted conditions, the differences were characterized by a seasonal trend, negative values were being more frequent at the beginning of the biomass burning season and positive values becoming dominant as the burning season progressed. A consistent seasonal pattern was also found in the variability of single scattering albedo and asymmetry parameter retrieved over Alta Floresta. We explored the hypothesis that the observed pattern of behavior in the irradiance differences may be linked to the modulation of the aerosol optical intensive properties by atmospheric humidity. Aerosol microphysical properties, specifically fine mode size parameters and





**Figure 8.** (a) Real and (b) imaginary part ( $k$ ) of the complex refractive index, (c) fine mode volume median radius ( $r_{fm}$ ) and (d) fine mode geometric standard deviation ( $\sigma_{fm}$ ) as a function of Column Water Vapor (CWV). Data are from sky retrievals products level 2.0 of the AERONET Sun photometer installed at Alta Floresta (1999–2007) and for aerosol optical depth larger than 0.4 at 440 nm.

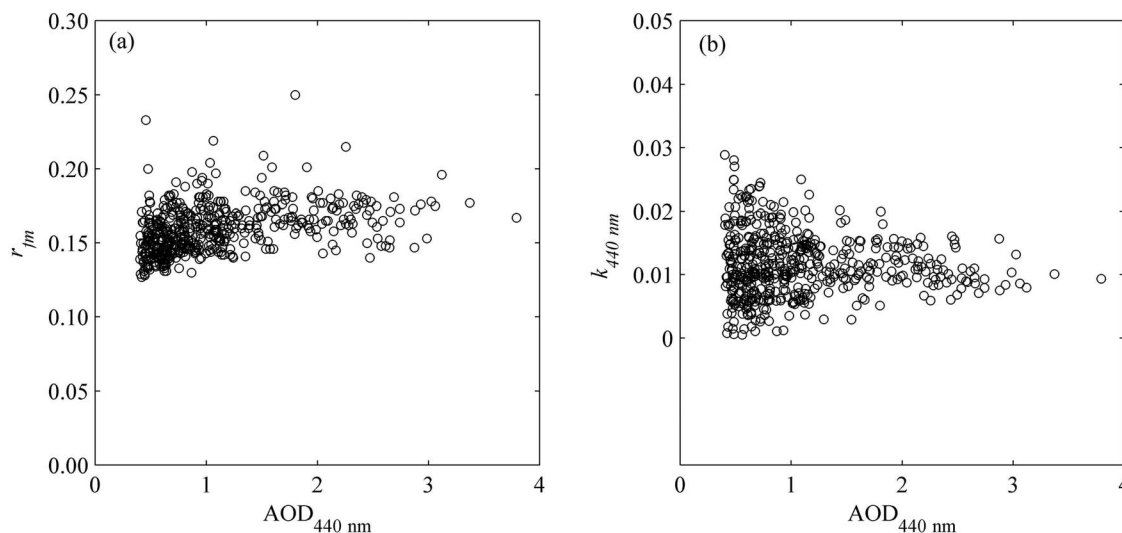
complex refractive index, were analyzed against atmospheric water vapor content. A sensitivity study using a Mie code was performed in order to assess the sensitivity of single scattering albedo and asymmetry parameter to the typical variations of each microphysical parameter over Alta Floresta. We found that the complex refractive index, specifically the imaginary part, explains most of the variability in  $\omega_{0\lambda}$ . Variations in fine mode size parameters, specifically volume median radius and geometric standard, had lower impact on  $\omega_{0\lambda}$ , mainly in the visible spectral region. As expected,  $g_{\lambda}$  is largely controlled by size parameters. The analysis of microphysical parameters as function of atmo-

sphere humidity showed that the imaginary part of the complex refractive index and fine mode volume median radius tend, respectively, to decrease and to increase as the amount of water vapor in the atmosphere increases. The more notable relation between  $k$  and column water vapor suggests that, if there is a causal relationship between humidity and aerosol intensive optical properties, that humidity-dependent processes that affect the chemical composition of aerosol particles are more likely to be responsible. However, despite the consistency of the trends observed for irradiance differences, atmosphere humidity and intensive properties of aerosols we are still not able to

**Table 2.** Impact of Using Typical Extreme Values of Fine Mode Distribution Parameters and Complex Refractive Index Retrieved Over Alta Floresta on Single Scattering Albedo ( $\omega_{0\lambda}$ ) and Asymmetry Parameter ( $g_{\lambda}$ ) in the Visible (VIS) and Near Infrared (NIR) Spectral Regions<sup>a</sup>

Microphysical Parameters	Typical Variability	Impact on $\omega_{0\lambda}$		Impact on $g_{\lambda}$	
		VIS (440 nm)	NIR (870 nm)	VIS (440 nm)	NIR (870 nm)
$r_{fm}(\mu m)$	0.12–0.18	<0.02	0.06	0.08	0.12
$\sigma_{fm}$	0.35–0.55	0.01	0.01	<0.01	0.07
$n$	1.35–1.55	0.04	0.07	0.05	<0.02
$k$	0.0015–0.02	0.08	0.12	<0.01	<0.01

<sup>a</sup>Distribution parameters are  $r_{fm}$ , volume median radius;  $\sigma_{fm}$ , geometric deviations. Complex refractive index is  $n + ik$ . The impacts were calculated using a Mie code and varying a specific parameter while keeping the others constant assuming their climatological values.



**Figure 9.** (a) Fine mode volume median radius ( $r_{fm}$ ) and (b) imaginary part of refractive complex index ( $k$ ) as a function of aerosol optical depth ( $AOD_{440\text{ nm}}$ ). Data are from sky retrievals products level 2.0 of the AERONET Sun photometer installed at Alta Floresta (1999–2007) and for aerosol optical depth larger than 0.4 at 440 nm.

be conclusive as to whether the trends are results of causalities. Nevertheless, the tie between the results from the analysis of two independent data sets highlights the potential role of humidity conditions in modifying aerosol intensive particles in the southern portion of the Amazon basin. Although our analysis focused on modeled downward irradiance at the surface, the implications of this issue extend to other applications, including retrieval of aerosol optical depth from space. Causal or not, the relationship between humidity variability and aerosol optical properties may be a useful tool which can be used to improve the climatological representation of aerosol intensive optical properties in the region, as has been done using aerosol loading [Remer *et al.*, 1998; Procopio *et al.*, 2004; Levy *et al.*, 2007]. For the Alta Floresta site in particular, during smoky conditions, there is a clearer relationship between certain microphysical parameters and CWV rather than with aerosol optical depth. An ongoing study is evaluating the ability of climatological optical models selected according to distinct humidity scenarios to reproduce the observed solar irradiance at the surface.

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H. Brindley, Physics Department, Imperial College London, Prince Consort Road, London SW7 2AZ, UK.

T. F. Eck and J. Schafer, Biospheric Sciences Branch, NASA, GSFC, Greenbelt, MD 20771, USA.

N. E. Rosário and M. A. Yamasoe, Atmospheric Sciences Department, University of São Paulo, Rua do Matao 1226, Butanta, São Paulo, SP 05508-090, Brazil. (nrosario@model.iag.usp.br)