Aerosol direct radiative forcing on Djougou (Northern Benin) during the AMMA dry season experiment (SOP_0).


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Abstract.

The purpose of this work is to investigate the direct radiative forcing of aerosols on the supersite of Djougou (Northern Benin) during the AMMA (African Monsoon Multidisciplinary Analyses) dry season experiment (January 2006). We focus our simulations on the top of atmosphere (TOA), bottom of atmosphere (BOA), and atmosphere (ATM) radiative forcings. During the period, sun-photometer measurements indicate a rather turbid atmosphere with a mean aerosol optical depth (AOD), for the overall period, about 0.90 ± 0.01 at 440 nm. The aerosol absorption coefficient estimated at the surface ranged between 2 and 90 Mm\(^{-1}\) (mean value 19.2 Mm\(^{-1}\) at 520 nm) and the scattering coefficient between 50 and 400 Mm\(^{-1}\) (mean 160 Mm\(^{-1}\) at 520 nm). This leads to a single scattering albedo (SSA) comprised between 0.75 and 0.95 (at 520 nm) with a mean value of 0.90, indicating moderate absorbing aerosols. In parallel, micro pulse lidar measurements indicate the presence of two distinct aerosol layers, with a first one located between the surface and 1 km and a second one located above (1.5 - 4.0 km). Based on surface and aircraft observations, sunphotometer measurements, lidar profiles and MODIS sensor, an estimation of the daily clear-sky direct radiative forcing has been estimated for the 17\(^{th}\) to 23\(^{th}\) January 2006 period. Simulations indicate that aerosols reduce significantly the solar energy reaching the surface (mean \(\Delta F_{\text{BOA}} = -64.2 \text{ Wm}^{-2}\)) by reflection to space (mean \(\Delta F_{\text{TOA}} = -19.0 \text{ Wm}^{-2}\)) but predominantly by absorption of the solar radiation into the atmosphere (mean \(\Delta F_{\text{ATM}} = +45.3 \text{ Wm}^{-2}\)). The mean heating rate at the surface and within the BBL is considerably enhanced by 1.48 and 1.91 °K by day, respectively.

1. Introduction

Although atmospheric aerosols are recognized to be a crucial agent of climate change [IPCC, 2007], the estimation of their impact on climate remains an important scientific challenge. Indeed, in contrast with the radiative forcing attributed to greenhouse gases, which may be determined to a reasonably high degree of accuracy, the uncertainties related to aerosol radiative forcings remain
very large [IPCC, 2007]. This is in the first place due to the complexity of aerosols. Indeed, atmospheric particles have different sources, sizes, chemical compositions, mixing, shapes, optical properties and lifetimes, which makes it difficult to get an accurate estimation of their climatic impact at regional or global scale.

Among all sources of atmospheric particles around the world, Western Africa is the world's largest source of biomass burning aerosols and mineral dust. Long past satellite observations indicate that these aerosol plumes, characterizing by high aerosol optical depths (often superior than one in the visible range) are the most widespread, persistent and dense observed at global scale. The effect of such plumes of dust and smoke aerosols on climate change represents one of the largest uncertainties in the Earth radiative budget.

In a first time, atmospheric aerosols emitted in Western Africa can modify the radiative budget directly by absorption or scattering of solar radiations. Secondly, they can modify clouds properties and/or lifetimes when particles act as Cloud Condensation Nuclei [Twomey, 1977]. Furthermore, as dust and smoke particles are able to absorb solar radiations, the radiative heating due to aerosol layers can modify the atmospheric dynamic and suppress convection, resulting in evaporation of clouds [Ackerman et al., 2000]. This effect, named “semi-direct” effect, can lead to a positive forcing by reducing cloud fractional cover and counterbalances the aerosol indirect effect. As an example, Koren et al. [2004] have clearly underlined the reduction in cumulus cloud cover over the Amazon region during the smoke season, whereas Abel et al. [2005] indicate significant regional direct effect of smoke particles over the Southern African region during anthropogenic burning activities.

Hence, dust and smoke particles coming from the West Africa region could strongly modify the regional or global climate as they have the potential to be exported over great distances by prevailing winds and atmospheric waves. A better representation of their climatic effects needs, in addition with modelling exercises and satellite observations, ground-based and integrated-column
measurements of their physical, chemical and optical properties, associated with their vertical repartitions. This specific question represents one of the objectives of the AMMA (African Monsoon Multidisciplinary Analysis) project [Redelsperger et al., 2007], as few data's characterizing optical properties and direct radiative forcing of aerosols in the Western Africa region are available in the literature.

In this work, we present results of aerosol optical properties and direct radiative impact obtained during January 2006, in the frame of the SOP_0 (Special Observation Period_0) experiment [Haywood et al., this issue], on the Djougou ground-based station (9.70N / 1.68W, Northern Benin), which was administered by the Laboratoire of Aerologie (France, Toulouse). Among all sites instrumented during AMMA, the measurements performed on Djougou represent an unique database for studying the microphysical and optical properties of expected mixed aerosols (dust and biomass burning), contrary to the other sites, more specifically deployed in the Sahelian dust transect, located northern of the main biomass burning areas (Figure 1). We focus our analysis during the dry season, as maximum emissions of biomass burning aerosols occur during the December to February period.

The estimation of the aerosol radiative forcing have been performed using the discrete ordinate Radiative Transfer Model (RTM) GAME (Global Atmospheric ModEl) [Dubuisson et al., 1996, 2004]. Radiative transfer computations depend on several parameters such as the spectral values of solar radiation incident on the atmosphere, the spectral variation of the aerosol optical depth (AOD), single scattering albedo (SSA), asymmetry parameter (g), surface albedo, associated with the vertical profiles of atmospheric (Temperature, Relative Humidity and ozone concentration) properties.

Over Djougou, all these informations have been obtained from ground-based measurements using sun-photometer, nephelometer, aethalometer, Differential Mobility Analyzer (DMA), Micro Pulse Lidar (MPL), UK Bae-146 aircraft measurements and MODIS (Moderate Resolution Imaging
Spectroradiometer) satellite observations. The methodology used for computing the aerosol radiative impact is described in detail by Mallet et al. [2006] and Roger et al. [2006] and are not repeated here. In this article, measurements of aerosol optical properties are presented in section 3 and the direct radiative forcing of aerosols obtained for the 17th-23rd January period are presented in section 4.

2. Experimental procedures

During the AMMA dry season experiment, several sites (Figure 1), (Djougou (Benin), Niamey (Niger), Banizoumbou (Niger) and M’Bour (Senegal)) were equipped with in situ and remote sensing instruments for characterizing dust and biomass burning aerosols microphysical and optical properties, their mixing, vertical repartition, transport and radiative forcings. These ground-based observations were completed by aircraft (Facility for Airborne Atmospheric Measurements BAe-146 from UKMO/NERC) and Ultra-light (LSCE/CNRS-CEA) measurements, as well as satellite (MODIS) observations. In this work, we used surface measurements performed at the station of Djougou, which was administered by the EDI team of the Laboratory of Aerologie (France, Toulouse). Hereafter, we do not detail the instrumentation dedicated for monitoring gas-phase components.

Aerosol ground-based instrumentation deployed at Djougou was dedicated to characterize the chemical composition of particles and their physical (number size distribution), chemical and optical (absorption and scattering) properties, by using, respectively, a 13-stage Dekati impactor, a DMA connected to a Condensation Particle Counter (TSI model 3010), a multi-channel Aethalometer (model AE-10, MAGEE Scientific; Berkeley, USA) and a single channel Nephelometer (model ECOTECH M9003). A pyranometer (Kipp & Zonen) has also been used to obtain the broadband (0.4-1.1 µm).

Associated with our ground-based instrumentation, a sunphotometer of the PHOTONS network
(French part of the AErosol RObotic NETwork, AERONET) was also used to estimate aerosol microphysical and optical properties for the whole atmospheric column [Dubovik et al., 2000a,b]. Finally, over Djougou, the vertical profiles of the attenuated backscattering and extinction coefficients have been estimated from an aerosol Micro Pulse Lidar (MPL) (model CE 370-2), developed by CIMEL, which is a micropulse system operating at 532 nm [Pelon et al., this issue]. Measurements are taken according to a standard sequence of 10 minutes acquisition every half an hour.

3. Aerosol Optical Properties

As mentioned in the introduction, radiative transfer computations depend in a first time on several aerosol optical parameters such as the spectral variation of the aerosol optical depth (AOD) (see 3.1), single scattering albedo (SSA) (see 3.2) and asymmetry parameter (g) (see 3.3).

3.1 Aerosol Optical Depth

Results of sun-photometer measurements obtained in January 2006 are presented in the Figure 2, where AOD at 440 nm (noted AOD$_{440}$), associated with uncertainty in AOD [Holben et al., 1998] are given. Although a much smaller amount of mineral dust has been observed during 2006, compared to previous years, Djougou atmosphere is rather turbid with AOD$_{440}$ exceeding 0.5 for major days. The mean AOD$_{440}$ during January 2006 is 0.90 ± 0.01, indicating a significant aerosol loading. Very high AOD are observed for the 17th and 18th January, with daily mean values of 1.5 and 1.1 (at 440 nm).

A way to know which kind of aerosols is to compute the Angström exponent [Angström, 1964], which represents the spectral variation of AOD. Indeed, this optical parameter (noted $\alpha$) furnishes an information of the aerosol type (averaged over the whole atmospheric column) and more specifically on its size and consequently on its anthropogenic or dust-like nature. For coarse aerosols, such as dust, $\alpha$ is close to 0 and for submicronic particles (such as biomass burning or
urban/industrial), αs comprised between 1 and 2. It can be computed from the following relation:

$$\alpha \lambda_1/\lambda_2 = - \log (\text{AOD} \lambda_1 / \text{AOD} \lambda_2) / \log(\lambda_1/\lambda_2)$$

(1)

In numerous studies, the Angström exponent is computed between 440 and 870 nm. Over Djougou, the mean value of $\alpha_{440/870}$ is $1.00 \pm 0.21$ over the total atmospheric column, revealing the presence of fine aerosols, expected to be due to biomass burning emissions.

3.2 Aerosol single scattering albedo

Two methods for estimating SSA at the surface are used in this study and summarized in the Table 1. The first one uses measurements of the scattering and absorption coefficients, with an AE10 aethalometer and ECOTECH nephelometer, providing SSA at one wavelength (520 nm), by using the following relation:

$$\text{SSA} = b_{\text{scat}} / (b_{\text{scat}} + b_{\text{abs}}) = b_{\text{scat}} / b_{\text{ext}}$$

(2)

The second method uses measurements of the aerosol size distributions together with the refractive index and the Mie theory [Mie, 1908]. This method, compared to the previous one, allows calculating SSA at different wavelengths required for radiative transfer modeling.

3.2.1 Determination of SSA from aethalometer and nephelometer measurements

3.2.1.1 Aerosol absorbing coefficient

The dry particle absorption coefficients (noted $b_{\text{abs}}$) was estimated by an AE10 aethalometer. It should be noted here that, at the surface, no corrections on optical properties due to Relative Humidity (RH) have been take into account as RH is lower than 30% for each days studied. The absorption coefficient is defined with the Bee-Lambert’s law:

$$I = I_0 \exp(-b_{\text{abs}} x)$$

(3)

where $I_0$ is the intensity of the incoming light and $I$ the remaining light intensity after passing through a filter with the thickness $x$. The attenuation (ATN) is given as a percentage values and is defined by the following relation:

$$\text{ATN} = \ln(I/I_0)$$

(4)
The aethalometer attenuation coefficient \( b_{\text{ATN}} \) is defined as:

\[
b_{\text{ATN}} = \frac{A}{Q} \times \left( \frac{\Delta b_{\text{ATN}}}{\Delta T} \right) \tag{5} \]

where \( A \) is the filter spot area, \( Q \) the volumetric flow rate and \( \Delta b_{\text{ATN}} \) is the change in attenuation during the interval \( \Delta t \). It is well known that \( b_{\text{ATN}} \) may differ from the true aerosol absorption coefficient \( b_{\text{abs}} \). Hence, the parameters \( C \) and \( R(\text{ATN}) \) are introduced by Weingartner et al. [2003] in order to convert aethalometer attenuation measurements to “real” absorption coefficients. The following relationship is used:

\[
b_{\text{abs}} = b_{\text{ATN}} \left( \frac{1}{C \cdot R(\text{ATN})} \right) \tag{6} \]

where \( C \) and \( R \) describe the two effects which modify the optical properties of filter particles. As mentioned by Weingartner et al. [2003], \( C \) represents corrections due to the multiple scattering of the light beam at the filter fibers, leading to enhance light absorption of the deposited particles [Lioussse et al., 1993]. The constant \( C \) is mainly depending on the nature of the filter used. Petzold et al. [1997] reported values of \( C \) comprised between 1.9 and 2.5. In this work, we used a value of 2.14 (Weingartner, personal communication).

The second effect, named “shadowing effect” is linked to the amount of aerosol particles embedded in the filter and optical properties of the deposited particles. This effect is described through the empirical function of \( R(\text{ATN}) \). For unloaded filters, \( R \) is equal to 1 (\( R(\text{ATN}=0) = 1 \)). In case of high filter loadings, the absorbing particles in the filter are able to absorb a higher fraction of the scattered light, leading to a reduction of the optical path in the filter (\( R \leq 1 \)).

Weingartner et al. [2003] reported that the “shadowing effect” of the particles is very pronounced for “pure” soot particles while it is almost negligible for aged atmospheric aerosols. This difference at the remote site is explained by the incorporation of a high amount of scattering aerosol material, which partially compensates the shadowing effect. Hence, in this work, considering that Djougou is not located at the sources of biomass burning emissions, we used \( R(\text{ATN}) = 1 \). Absorption coefficient is then computed as follow:
The dry absorption coefficient measured at 520 nm ($b_{\text{abs}(520)}$) is represented in the Figure 3. Results show that $b_{\text{abs}(520)}$ ranged from 2 to 90 Mm\(^{-1}\) (a megameter 1Mm\(^{-1}\) = 10\(^4\)m\(^{-1}\)), with an average for the overall period of about 19.2 Mm\(^{-1}\). Figure 3 indicates significant values of $b_{\text{abs}(520)}$ for the 17\(^{th}\)-19\(^{th}\) January period (~ 40 Mm\(^{-1}\)). The average value is comparable to $b_{\text{abs}}$ values obtained over the Indian Ocean during INDOEX (16 ± 8 Mm\(^{-1}\) from Mayol-Bracero et al. [2002]) and significantly higher to values reported during TARFOX (average of 3 ± 3 Mm\(^{-1}\) by Hegg et al. [1997]), ACE-Asia for polluted aerosols (8.6 Mm\(^{-1}\) from Anderson et al. [2003]), ACE-2 (8.9 Mm\(^{-1}\) from Öström and Noone [2000]), or in urban/industrial atmosphere during the ESCOMPTE experiment (5.6 – 9.3 Mm\(^{-1}\) from Mallet et al. [2005]).

3.2.1.2 Aerosol scattering coefficient

The dry particle scattering coefficients (noted $b_{\text{neph}(520)}$) was measured at 520 nm, between the 15\(^{th}\) and 30\(^{th}\) January 2006, by an ECOTECH nephelometer. Air is sampled through the semi-isokinetic inlet and the sample relative humidity is maintained below 40 %. The time resolution of the scattering coefficient measurement is about 1 second. One of the major sources of errors from the nephelometer originates from the instrument geometrical limitations as the nephelometer can not detect the entire phase function of the scattered light. It only measures aerosol light scattering coefficient for angular integration of 7°-170°. Since the light scattered by the particles within the measurement chamber is only integrated between 7° and 170°, the results provided by the nephelometer measurements underestimate the actual total scattering coefficients. This effect has been shown to be low for submicronic ($D_p \leq 1 \mu$m) aerosols, whereas the errors can be higher [Alfaro et al., 2003; Heintzenberg et al., 2006] for supermicronic ($D_p \geq 1 \mu$m) particles. In order to take into account this effect, the following relation is often used:

$$b_{\text{scat}(520)} = b_{\text{neph}(520)} \cdot C_{\text{corr}}$$  \hspace{1cm} (8)
where \( b_{\text{neph}(520)} \) is the dry scattering coefficient directly measured from the nephelometer, \( C_{\text{corr}} \) is the correction factor and \( b_{\text{scat}(520)} \) is the real dry scattering coefficient.

For anthropogenic aerosols, Carrico et al. [2000] reported a slight underestimation of the total scattering coefficient about \( \sim 4\% \) and Formenti et al. [2002] reported a total correction factor (\( C_{\text{corr}} \)) ranging between 1.08 and 1.05 (450 and 700 nm) for biomass burning aerosols, characterized by a modal diameter around 0.15 mm. Finally, Mallet et al. [2005] reported \( C_{\text{corr}} \) of 1.10 for urban/industrial submicronic aerosols.

In numerous studies, \( C_{\text{corr}} \) is usually estimated from the wavelength dependence of light scattering, as measured by the TSI 3\( \lambda \) nephelometer, based on Anderson and Ogren [1998]. In case of a single channel nephelometer, as the ECOTECH one used in our study, the spectral dependence of scattering is unavailable. Consequently, in order to correct scattering coefficients over Djougou, an alternative method has been adapted. In a first time, the aerosol size distribution has been estimated from DMA (Differential Mobility Analyser) analyses. Then, a correction factor deduced from the literature has been applied.

Here, the aerosol number size distribution was determined using a Vienna type medium size DMA, connected to a Condensation Particle Counter (TSI model 3010) at ambient RH every 15 minutes. It should be recalled here that, due to the low RH (around 20%) at the surface during the SOP_0, we consider aerosol microphysical and optical properties as dry during this period. Results of a few representative number size distributions, averaged for one day, are reported in the Figure 4. We can clearly observe that number size distributions are monomodal during the dry season, with the presence of a submicronic accumulation mode around 100 nm. It should be noted here that a good agreement (Figure 5) has been found with Passive Cavity Aerosol Spectrometer Probe (PCSAP) measurements performed on board the UK BAe-146 aircraft for the passage over Djougou (altitude of 500 m) for the January 19th.

As the number size distributions of aerosols over Djougou may be considered as mainly
submicronic, the corrections which are needed on nephelometer measurements are expected to be low. Then, we considered a correction factor ($C_{cor}$) of 1.05 in our computations following Formenti et al. [2002]. The dry scattering coefficient estimated at 520 nm is represented in the Figure 6. The values from our measurements ranged from 50 to 400 Mm$^{-1}$, with a mean value, for the overall period, of about 160 Mm$^{-1}$. These values are found to be significantly higher than those obtained during different field experiments, such as TARFOX (average of 44 ± 50 Mm$^{-1}$, Hegg et al. [1997]), INDOEX (62 ± 34 Mm$^{-1}$, Mayol-Bracero et al. [2002]), ACE-Asia (55.3 Mm$^{-1}$, Anderson et al. [2003]), ACE-2 (40 ± 17 Mm$^{-1}$, Quinn et al. [2000]) or ESCOMPTE (35 – 63 Mm$^{-1}$, Mallet et al. [2005]).

3.2.1.3 Estimation of SSA from aethalometer (3.2.1.1) and nephelometer (3.2.1.2)

The temporal variation of dry SSA$_{520}$ is reported in Figure 7. We can observe values comprised between 0.75 and 0.99, indicating high heterogeneities in SSA during the SOP_0. The mean value, for the overall period, is 0.90, indicating moderate absorbing particles. Figure 7 indicates that aerosols are mostly absorbing (SSA of 0.85) during the 17th-19th period, and mostly scattering (SSA of 0.95) for the 20th-25th period. It should be mentioned here that a good agreement has been observed, for the January 19th, between SSA estimated at the surface (SSA$_{520} = 0.89$) and the one measured onboard the UK BAe-146 (Figure 8) aircraft (SSA$_{520,BAe} = 0.90$). This result indicates a good level in confidence concerning our optical measurements and especially on the scattering correction used in the relation 8.

3.2.2 Determination of SSA from aerosol size distribution and assumed refractive index

In order to access to the spectral variation of SSA, calculations with a Mie code have been performed from the aerosol number size distribution measured with DMA (see above) and the refractive index of aerosols.

3.2.2.1 Characteristics of the Aerosol number size distribution

The aerosol number size distributions measured with the DMA (Figure 4) have been fitted by a
lognormal function as shown in the Figure 9 for the January 19th. According to Putaud et al. [2000], the equivalent mobility diameter as measured by the DMA was considered to be equal to the geometric diameter, $D_p$. The main characteristics (number of particles, Geometric Mean Diameter (GMD) and Geometric Standard Deviation (GSD)) of the lognormal functions, obtained for each studied days, are reported in the Table 2 (the data's are not available for the 20th January). The averaged values indicate a total number concentration of 6100 $\text{#.cm}^{-3}$, a GMD of 96.5nm, and a GSD of 1.87.

These characteristics are in the same order of values obtained by Rissler et al. [2006] in case of smoke particles produced over the Amazonian region during the dry season (GMD of 92 nm for the accumulation mode, associated with a GSD of 1.63). In terms of aerosol concentrations, Rissler et al. [2006] also reported a value of 5213 $\text{#.cm}^{-3}$ for the accumulation mode. In the compilation by Reid et al. [2005], median diameters between 100 and 130 nm are reported for smoke from grass, savanna, cerrado and forest burning. Finally, during the SMOCC field experiment, airborne measurements of particle size distribution [Guyon et al., 2005] have showed GMD of 110 ± 15 nm in 69 plumes within the boundary layer.

3.2.2.2 Estimation of Aerosol Refractive Index

In addition to the aerosol number size distribution, the Mie theory requires the knowledge of the refractive index of particles. This information is not available from our measurements. Therefore, over Djougou, the imaginary part of the refractive index has been obtained by constraining SSA computated with the Mie theory with SSA estimated using simultaneous measurements of absorption and scattering coefficients of particles (see the previous paragraph). In our computations, the real part of the refractive index is fixed to 1.54 in the visible range. It should be noted that the real part is almost similar for biomass burning in Africa (1.54) [Haywood et al., 2003] and mineral dust (1.55) [Dubovik et al., 2002]. As reported in the Table 2, the imaginary part of the refractive index is comprised between 0.004 and 0.035, with a mean of 0.015 (at 520 nm). For comparison, the
mean value obtained over Djougou is slightly lower than those reported by Haywood et al. [2003] (0.018 at 550 nm) and Dubovik et al. [2002] (0.021) for aged biomass burning plumes and smoke from African savanna, respectively. This could be due to the presence of dust in the boundary layer over Djougou, which can decrease the imaginary part of the refractive index.

3.2.2.3 Estimation of SSA from number size distribution (3.2.2.1) and refractive index (3.2.1.2)

Once obtained aerosol size distribution and refractive index, SSA has been computed at the seven wavelengths (Table 1) used in GAME. The values reported in Table 1 show a minor decreasing of SSA with the wavelengths (SSA$_{440}$ = 0.92 / SSA$_{870}$ = 0.90), results that are similar to those of Haywood et al. [2003] (SSA$_{450}$ = 0.91 / SSA$_{700}$ = 0.88) for smoke particles during SAFARI 2000 (South Africa biomass burning). It should be remarked that comparisons with SSA estimated from AERONET indicate significant differences (Table 1, Figure 8), with AERONET retrievals generally smaller. Such differences are due to the vertical structure of particles over Djougou, which are not homogeneous as shown by the lidar profiles (Figure 10). Furthermore, we can note that SSA spectral variations differ between the two methods, with an SSA increase with wavelengths from AERONET (SSA$_{440, AERO.}$ = 0.84 / SSA$_{870, AERO.}$ = 0.87). This is expected to be due to the differences in the aerosol size distribution, which is considered as monodal at the surface (Figure 4) and bimodal (fine and coarse modes) from AERONET retrievals.

3.3 Aerosol asymmetry parameter

Over Djougou, g has been computed from the Mie theory following the same methodology described for SSA. As shown in the Table 3, our results are included in the 0.59 and 0.63 range (at 520 nm) with a mean of 0.61 ± 0.01. As observed for SSA, g is decreasing with the wavelength (g$_{440}$ = 0.64 / g$_{870}$ = 0.53), due to the monomodal number size distribution (Figure 4) used in the Mie theory. Furthermore, as noted for SSA, some differences appear between our surface computations and AERONET retrievals, especially at longer wavelengths (870 and 1020 nm). In most of the cases, g retrieved for the whole atmospheric column is larger than the one computed at the surface (Table
3). As concluded for SSA, this is certainly due to differences in the aerosol size distribution. Indeed, the presence of the coarse mode in the AERONET aerosol size distribution tends to increase $g$.

4. Aerosol Radiative Forcing

In this study, we have computed the direct radiative forcing for January 17th to 23th. This period is pertinent due to the large range of AOD ($0.8 \leq \text{AOD (at 440 nm)} \leq 1.5$) and SSA ($0.83 \leq \text{SSA (at 520 nm)} \leq 0.98$, see Table 1). All the results of the simulations are summarized in Table 4 and in Figure 11.

4.1 Radiative Transfer Modeling

More details on the methodology employed for computing the diurnally averaged clear sky radiative forcing are given in Mallet et al. [2006] and Roger et al. [2006] and are not recalled here. The clear-sky direct radiative forcing over Djougou has been estimated from the GAME radiative transfer model, detailed in Dubuisson et al. [2004]. GAME accounts for the scattering and absorption processes by particles and gases. Gaseous absorption ($\text{H}_2\text{O}, \text{CO}_2, \text{O}_2$ and $\text{O}_3$) is treated from the correlated k-distribution, using a line by line code [Dubuisson et al., 1996] and multiple scattering effects are treated using the Discrete Ordinates Method (DOM) [Stamnes et al., 1988]. This method allows accurate treatment of scattering and absorption by aerosols, clouds and molecules.

Upward and downward net radiative fluxes are calculated over the spectral solar, ranging from 2500 to 50000 cm$^{-1}$, with a 100 cm$^{-1}$ spectral resolution. Calculations of radiative fluxes integrated over the entire shortwave region are performed at every 1 hour interval and for 24 hours period. From these fluxes, we computed the aerosol clear-sky daily direct forcing at the bottom of atmosphere (BOA), $\Delta F_{\text{BOA}}$, and at the top of the atmosphere (TOA, 70 km in this case), $\Delta F_{\text{TOA}}$. The first one represents the effect of particles on the net short-wave radiation fluxes reaching the surface and the second one the radiation fluxes reflected back to space by aerosols. BOA and TOA forcings
have been calculated as following:

\[ \Delta F_{\text{BOA}} = F_{\text{BOA}(w)\downarrow} - F_{\text{BOA}(0)\downarrow} \]  
\[ \Delta F_{\text{TOA}} = - (F_{\text{TOA}(w)\uparrow} - F_{\text{TOA}(0)\uparrow}) \]  

where \( F_{\text{BOA}(w)\downarrow} \) and \( F_{\text{BOA}(0)\downarrow} \) are, respectively, the downward net radiative fluxes simulated at the surface with (w) and without (0) aerosols. \( F_{\text{TOA}(w)\uparrow} \) and \( F_{\text{TOA}(0)\uparrow} \) are, respectively, the upwards net radiative fluxes simulated at the top of atmosphere with (w) and without (0) aerosols. With this convention, a negative sign of \( \Delta F \) implies an aerosol cooling effect. Finally and as reported in Roger et al. [2006], we compute the atmospheric forcing, \( \Delta F_{\text{ATM}} \), by using the following relation:

\[ \Delta F_{\text{ATM}} = \Delta F_{\text{TOA}} - \Delta F_{\text{BOA}} \]  

\( \Delta F_{\text{ATM}} \) represents the possible absorption of solar radiations due to absorbing particles within the atmospheric layer where aerosols are located.

### 4.2 Aerosol Vertical Profiles

As reported by Ganguly et al. [2006], an important point for estimating the direct forcing of aerosols is their vertical profiles within the atmosphere. Indeed, numerous studies [Haywood and Ramaswamy, 1998; Chung et al., 2005] indicate that one major contributor to the uncertainty in atmospheric forcing comes from uncertainty in the vertical distribution of aerosols. Differences in vertical profiles affect the distribution of forcing within the atmosphere and can produce different patterns in terms of cooling or heating as the function of the altitude.

#### 4.2.1 Vertical profiles of Aerosol Extinction Coefficient

In order to take into account the extinction vertical profiles in GAME, we have used a MPL, operating on Djougou and administered by the Service of Aeronomie (SA). Once corrected from signal saturation and geometrical factor, the attenuated profiles are inverted in order to retrieve extinction coefficient profiles, \( \alpha_e \) at 532 nm [Pelon et al., this issue]. Figure 10 shows an example of extinction coefficient (in km\(^{-1}\)) profiles obtained at Djougou for January 21\(^{th}\) (the shape of the
vertical profile is similar to those obtained during all the SOP_0, presented in Pelon et al. [this issue]). The boundary layer height (BLH) is around 0.5 km and characterized by extinction coefficients around 0.15 km\(^{-1}\), following by a rapid decrease of \(\alpha_e\) between 0.5 and 1 km. Above 1 km, we can observe a second aerosol layer, with \(\alpha_e\) of 0.3 km\(^{-1}\), for altitudes comprised between 1500 and 4000 m, assumed to be mainly composed by biomass burning aerosols (Osborne, personal communication). MPL measurements show clearly the significant contribution of the biomass burning layer (BBL) to the total AOD (around 70%) and the more negligible contribution of the BHL to total AOD (around 30%), indicating that a large part of the solar extinction over Djougou is due to the BBL.

4.2.2 Vertical profiles of Aerosol SSA and g

Associated with the aerosol extinction profiles, SSA and g vertical profiles were obtained by combining our measurements at the surface, with aircraft observations and literature data's. Briefly, the spectral variation of SSA and g computed at the surface (Table 1 and 3) are used as input for the BLH. Concerning the BBL, we considered the spectral variation of dry SSA and g obtained onboard the UK Bae-146 by Jonhson et al. [this issue] for biomass burning aerosols. Finally, for free troposphere and stratosphere, as reported by Roger et al. [2006], the spectral variation of optical properties coming from the aerosol models described by Hess et al. [1998] has been used.

As reported in the part 3, no corrections on optical and microphysical properties due to RH have been taking into account in the BHL as RH is lower than 30% for each studied days. However, RH higher than 50% has been observed in the BBL, as shown on the Figure 12 (for January 19\(^{th}\)). In such case, dry optical properties need accurate corrections to take into account variations with RH, especially on scattering. In our computations, the corrections are only performed for January 18\(^{th}\) and 19\(^{th}\). The RH dependence on \(b_{scat}\) has been fitted with the following empirical function:

\[
b_{scat} = b_{scat, dry} \left[1 + a(RH/100)^b\right]
\]

where \(b_{scat}\) is the light scattering at ambient RH, \(b_{scat, dry}\) is the light scattering coefficient in dry
state, a and b are empirical fitting parameters. In this study, the values of a and b (a = 1.31 ± 0.08 and b = 4.88 ± 0.28) are taken from Magi and Hobbs [2003] for ambient smoke samples. Ambient SSA has been corrected with new b\text{scat} values. As an example of results, for January 19th, dry to wet SSA changes from 0.86 to 0.90 (at 450 nm).

Finally, in order to correct the asymmetry parameter with RH in the BBL, we have used in the Mie code the microphysical properties (GMD = 0.24 μm / GSD = 1.30) and refractive index (1.54 - 0.035i) reported in dry state by Johnson et al. [this issue] for biomass burning particles. GMD has been calculated in the ambient state from the Growth Factor (GF) reported by Rissler et al. [2006], which corresponds to moderately hygroscopic biomass burning aerosols observed in South America (during dry season). To finalize Mie computations, the aerosol refractive index has been computed at ambient RH (for 18th and 19th) using the relation described in Mallet et al. [2003]. As an example of results, Mie computations indicate a change in g from 0.58 (at 550 nm) in dry state to 0.65 at 85% RH for January 19th.

4.3 Vertical profiles of atmospheric properties

RH and temperature vertical profiles used in GAME have been measured onboard the UK BAe-146 aircraft for January 19th. We completed the stratification, for altitudes larger than 4 km, by using data's available in the Shadoz network [Thompson et al., 2003]. Ozone concentration profiles have been determined by combining ground-based measurements performed at Djougou for the BHL and Shadoz [Thompson et al., 2003] measurements carried out on Cotonou (Southern Benin) for the upper atmosphere.

4.4 Surface albedo

As the estimation of radiative forcing has been performed over land, surface reflection is an issue. As reported by Roger et al. [2006], we used the MODIS albedo product (MODIS/Terra Albedo; 16-Day; Level-3 Global 1 km SIN Grid), which provides the spectral dependence of surface albedo at seven wavelength bands. Such informations have been used as input in GAME to reproduce the
surface albedo for the entire shortwave range. In our computations, a constant surface albedo has been considered for the whole month of January 2006. At 550 nm, the surface albedo at Djougou is equal to $0.20 \pm 0.06$.

### 4.5 Bottom of atmosphere Direct Radiative Forcing

In a first time and as reported in Figure 13, the downwards fluxes simulated at the surface by GAME (uncertainties of $\pm 15\%$, see Roger et al. [2006]) are well compared with pyranometer measurements performed at Djougou, with a mean difference of $\pm 5\%$ for the period investigated. As summarized in the Table 4, simulations indicate important surface radiative forcings for each studied days, with $\Delta F_{\text{BOA}}$ always negative ($\Delta F_{\text{BOA}}$ values are comprised between $-54.1$ and $-85.1$ W.m$^{-2}$; mean $-64.2$ W.m$^{-2}$). This simulated daily radiative forcing over Djougou is found to be higher than those reported during previous experimental campaigns on anthropogenic aerosols, such as TARFOX ($-26$ W.m$^{-2}$, Hignett et al. [1999]) or INDOEX ($-45$ W.m$^{-2}$, Ramanathan et al. [2001]; Leon et al. [2002]). The difference is mainly due to higher AOD obtained over the region of fires during the period investigated compared to previous campaigns.

Our simulations allow investigating the surface forcing during the 17th and 18th January, with high AOD (1.5 at 440 nm). For these specific days, the instantaneous surface forcing estimated by GAME can reach up to $-240$ W.m$^{-2}$ and $-165$ W.m$^{-2}$, for January 17th and 18th, respectively (for a solar zenith angle of 30°). Such values are found to be very consistent with observations reported by Schafer et al. [2002] for biomass burning aerosols in the Zambian Savanna (AOD = 1), with a reduction in total irradiance at the surface of 210 W.m$^{-2}$ (mean solar zenith angle of 31 degrees).

It is clear that such significant decrease of solar energy at the surface can strongly modify the surface energy budget. Indeed, under normal conditions, surface radiation flux is balanced by evaporation (latent heat fluxes) and sensible heat flux from the surface. In order to compensate for the reduction of surface solar flux, a decrease of heat fluxes released from the surface may be expected. Feingold et al. [2005] or Jiang and Feingold [2006] have showed that the reduction in
surface latent and sensible heat fluxes associated with biomass burning impact may reduce cloudiness over the Amazonia region. Consequently, and due to our high BOA values, it appears essential to study the biomass burning impact on the surface energy budget, especially in terms of latent and sensible fluxes.

4.6 TOA and Atmospheric Direct Radiative Forcing

GAME calculations of upwards fluxes have been compared with Broad Band Radiometer (BBR) flux measurements performed on board the BAe-146 aircraft for January 19th and satisfactory agreement have been obtained between the normalised upwards fluxes simulated with GAME (146 W.m\(^{-2}\)) and the one measured from BBR (140 W.m\(^{-2}\)), for a solar angle of 30° (12:00 UTC). Over Djougou, moderate or low $\Delta F_{\text{TOA}}$ (Table 4) have been computed by GAME, with values comprised between -14.8 W.m\(^{-2}\) and -23.7 W.m\(^{-2}\) (mean value -18.9 W.m\(^{-2}\)). The higher value of $\Delta F_{\text{TOA}}$ is obtained for the January 22nd, corresponding to higher SSA (0.98, Table 1) of particles in the BHL, which reflect more efficiently the solar radiations back to space.

One of the most important results of this study concerns the large atmospheric forcings obtained over Djougou due to significant differences between $\Delta F_{\text{TOA}}$ and $\Delta F_{\text{BOA}}$. Indeed, our results indicated that $\Delta F_{\text{TOA}}$ is 3.5 times lower than the mean $\Delta F_{\text{BOA}}$. During the SOP_0, $\Delta F_{\text{ATM}}$ ranges from 36.6 to 61.5 W.m\(^{-2}\) (mean value of 45.3 W.m\(^{-2}\)). As observed for BOA, maximum $\Delta F_{\text{ATM}}$ is obtained for January 17th, due to high AOD (1.5) and low SSA (0.87).

As mentioned by Ganguly et al. [2006], although the large difference between the BOA and TOA forcing may indicate the trapping of solar radiations in the atmosphere, it is of great importance to investigate how this atmospheric heating is vertically distributed within the atmosphere. Indeed, several studies have shown that the atmospheric heating by absorbing particles may evaporate low-level clouds, resulting in a decrease of cloud cover and planetary albedo [Hansen et al., 1997; Ackerman et al., 2000]. For that purpose, vertical profile of the HEating Rate (HER in °K by day) has been computed from GAME, as described by Roger et al. [2006]. The average vertical profile of
the predicted HER, due to the only presence of aerosols, is reported on the Figure 14 and the corresponding mean HER values, for each studied days and for the two main aerosol layers (BHL & BBL), are indicated in the Table 4.

Figure 14 shows that HER decreases very sharply above two hundred meters to reach low values at 1 km. Above 1 km, the aged biomass burning aerosol considerably increases the average heating rate in the layer between 1500 and 4000 m altitude. Simulations indicate that HER due to aerosols is comprised between 0.66 and 2.20 °K by day (mean value 1.48 °K by day) in the BHL (Table 4) and between 1.29 - 3.27 °K by day in the BBL (mean of 1.91 °K by day). HER in the BBL reaches up to 3.27°K by day for January 17th. For comparison, Keil and Haywood [2003] observed that biomass burning aerosols increase HER by 1.77 °K by day in the BBL (1.8 - 3.7 km) during SAFARI 2000. Authors indicate also that HER can reach up to 2.05 °K by day for SSA of 0.86. These values are found to be very consistent with the one obtained from our GAME simulations (1.91 °K by day) over Djougou.

5. Conclusion

The purpose of this work is to investigate the clear-sky direct radiative forcing of aerosols over the ground-based station of Djougou, located in Northern Benin, during the AMMA (African Monsoon Multidisciplinary Analyses) dry season experiment. The results presented in this work are based, in a first time, on ground-based measurements for estimating aerosol optical (scattering and absorption coefficients) and physical (number size distribution) properties. Additionally, we used UK Bae-146 aircraft, PHOTON sun-photometer, micro pulse lidar and MODIS observations for estimating, respectively, vertical profiles of thermodynamic parameters, spectral variation of the aerosol optical depth (AOD), aerosol extinction vertical profiles and surface albedo. All these informations have been used as input in the discrete ordinate radiative transfer model GAME, for computing the clear-sky direct radiative forcing of aerosols during the 17th - 23rd January period.
During January 2006, sun-photometer measurements indicate a mean AOD of 0.90 ± 0.01 at 440 nm, indicating a significant aerosol loading over Djougou. The average aerosol absorption and scattering coefficients at the surface are 19.2 Mm\(^{-1}\) and 160 Mm\(^{-1}\) (at 520 nm), respectively, leading to a mean single scattering albedo of 0.90 (moderate absorbing particles) over the period studied. In the same time, the aerosol number size distribution is shown to be clearly monomodal, with an accumulation mode, characterized by a mean geometric diameter of 96.5 nm and a geometric standard deviation of 1.87.

In addition to ground-based observations, vertical profiles of aerosol extinction coefficient have been investigated through a micro pulse lidar (MPL). Our results indicate a complex vertical structure, characterized by the presence of two aerosol distinct layers, with a first one located between the surface and 1 km and a second one located above (1.5 – 4.0 km). Furthermore, lidar profiles indicate also that the majority of the solar extinction over Djougou is due to the second aerosol layer, composed in majority by smoke particles, which contribute to 70% of the total AOD.

Our radiative transfer simulations indicate that aerosols over Djougou lead to a significant diurnal average reduction of 64.2 W.m\(^{-2}\) in the surface solar radiation, an increase of 45.3 W.m\(^{-2}\) in the atmospheric solar absorption, and an increase of 18.9 Wm\(^{-2}\) in the reflected solar radiation at the top of atmosphere. Our simulations show also that the instantaneous surface forcing predicted by GAME can reach up to significant values of -240 W.m\(^{-2}\) for January 17\(^{th}\) (AOD of 1.5).

Changes in heating rates due to the presence of aerosols have been also investigated and discussed. The results we obtained indicate that the heating rates within the BHL and BBL are considerably enhanced, with a mean value of 1.48 and 1.91 °K by day, respectively. In regards to such radiative forcings and changes in heating rates, future works are now clearly required for investigating the possible impact on the surface-atmosphere system during the dry season over the West African region.
Acknowledgments

Based on a French initiative, AMMA was built by an international scientific group and is currently funded by a large number of agencies, especially from France, UK, US and Africa. It has been the beneficiary of a major financial contribution from the European Community's Sixth Framework Research Programme. AMMA has been endorsed by IGBP (IGAC, ILEAPS) and WCRP (GEWEX, CLIVAR). Detailed information on scientific coordination and funding is available on the AMMA International web site (http://www.amma-international.org). We would like also to thank the PHOTON staff for the data collection, calibration, and processing and especially the PHOTON PIs: D. Tanre (LOA) and L. Blarel (LOA). FAAM is jointly funded by the Met Office and the Natural Environment Research Council.
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Table 1. Aerosol Single Scattering Albedo (averaged between 08:00 – 18:00 UTC) estimated for different days on Djougou at the surface (at 520nm) from Nephelometer and Aethalometer, at the surface from the Mie theory (at different wavelengths) and for the whole atmospheric column from AERONET retrievals.
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Table 2. The number concentration, Geometric Mean Diameter (nm), Geom. Standard Dev. for the lognormal distribution, and the refractive index (at 520nm) of particles over Djougou.
Table 3. Aerosol asymmetry parameter estimated at the surface (at different wavelengths) from the Mie theory and for the whole atmospheric column (from AERONET retrievals).

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Table 4. Aerosol Optical Depth at 440 nm, daily clear-sky direct bottom of atmosphere (BOA), top of atmosphere (TOA), atmospheric (ATM) radiative forcings (in W.m\(^{-2}\)), heating Rates (HER, in °K by day) only due to aerosols in the BHL (averaged between 0 and 1000 m) and in the BBL (averaged between 1500 and 4000 m).

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Figures.

Figure 1. Localisation of the Djougou ground-based station (Northern Benin).

Figure 2. Aerosol optical depth (AOD) retrieved at 440 nm from sun-photometer measurements (PHOTON / LOA) over Djougou during January 2006.

Figure 3. Dry aerosol absorbing coefficient ($b_{abs,520}$) retrieved at 520 nm from the AGEE Aethalometer.

Figure 4. Dry aerosol number size distribution measured from SMPS on Djougou.

Figure 5. Comparisons between number aerosol size distributions measured over Djougou and onboard the UK BAe-146 aircraft for the passage over Djougou (19th January).

Figure 6. Dry aerosol scattering coefficient ($b_{scat,520}$) measured at 520nm and at the surface on Djougou during the 17th to 30th January period.

Figure 7. Dry aerosol single scattering albedo ($SSA_{520}$) estimated at the surface (at 520nm) during the 17th to 30th January period.

Figure 8. Comparisons between SSA measured at the surface, onboard the aircraft and retrieved from AERONET.

Figure 9. Aerosol size distribution measured from the SMPS (♦) and fitted by a lognormal function (—) for the 19th January.

Figure 10. Micro Pulse LIDAR measurements obtained over Djougou for the 21th January 2006.

Figure 11. Aerosol clear-sky daily direct radiative forcing (in W.m$^{-2}$) estimated for the 17th - 23th January period.

Figure 12. Relative humidity profiles obtained from the BAE-146 aircraft during the 19th January.

Figure 13. Observed versus modeled surface global fluxes over Djougou. The solid line corresponds to perfect agreement.

Figure 14. Mean vertical profiles of the predicted Heating Rate due only to the presence of aerosols during the 17-23th January.
Figure 1.
Figure 2.
Figure 3.
Figure 4.
Figure 5.
Figure 6.
Figure 7.
Figure 8.
Figure 9.
Figure 10.
Figure 11.
Figure 12.
Figure 13.
Figure 14.