

A STUDY ON THE IMPACTS ON CLIMATE CHANGE DUE TO THE INCREASE IN THE CONCENTRATION OF ANTHROPOGENIC AEROSOLS

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Abstract. *In recognition of the growing influence of the aerosol effect in the local/regional climate change, in this work the aerosol properties are studied in order to estimate the direct aerosol radiative forcing. The aim of this paper is to call the attention for the biomass burning aerosols in the Brazilian Amazon and cerrado regions. Biomass burning is an important source of carbonaceous aerosols over the tropical rainforest. Various studies have showed that there is a large uncertainty in the evaluation of the aerosol direct radiative forcing due to incomplete knowledge of global aerosol properties and their temporal changes. Two radiative models are used to estimate direct forcing at the top of the atmosphere, at the Earth's surface and within the atmosphere. The results suggested that the increase in concentration of biomass burning aerosols may cause opposite effects: a cooling and a warming effect.*

Keywords. *Biomass burning, Direct radiative forcing, Aerosol properties, Radiative models*

1. Introduction

Anthropogenic aerosols emissions to the atmosphere have increased substantially over the past 156 years as a consequence of the Industrial Revolution. Aerosols are an important constituent of the atmosphere which has several environmental impacts. Their effects include: (1) human adverse health effects; (2) reduce visibility associated with haze; (3) scattering and absorption of incoming solar radiation (direct effect); and (4) modifying the microphysical and optical properties of clouds (indirect effect). Both, direct and indirect effects cause perturbations in the Earth's radiation budget.

The relationship between the human health effect and airborne aerosols can be related with the size of the particles (Barat et al., 2002). Coarse mode aerosols, ($2.5 < d_{ap} < 100 \mu\text{m}$, where d_{ap} is the aerodynamic diameter), may penetrate in the lung and aggravate diseases such as asthma (Barat et al., 2002). Fine mode aerosols, ($0.001 < d_{ap} < 2.5 \mu\text{m}$), are associated with respiratory diseases and mortality. These fine particles deposit in the alveolar region of the lungs and can contribute to health deterioration. They also can be absorbed into the lung and cause alterations of lung tissues and a decrease of the pulmonary function. Visibility degradation reflects the attenuation of solar radiation in the atmosphere and arises from: (1) reduction in contrast between object and the background; and (2) attenuation of the light signal from the object due to the absorption and scattering of light caused by pollutant gases and also by fine particles in the range of $0.1 < d_{ap} < 1.0 \mu\text{m}$ (Chan et al., 1999).

Atmospheric aerosols can act as cloud condensation nuclei (CCN), which may have an indirect effect on the climate system by changes in the microphysical (e.g. droplet number), micro-chemical, radiative and optical properties of clouds (first indirect effect, Twomey 1974; Twomey, et al., 1984; Schwartz, 1996; IPCC 2001). The enhanced number of CCN may alter nucleated droplet number concentrations. As cloud droplet number concentrations increase, for constant liquid water content, the mean droplet size decreases, which will increase the cloud's albedo, giving a negative radiative forcing. The enhanced droplet radius might also reduce the precipitation from clouds, modifying cloud cover and cloud lifetime (second indirect effect, Lohmann and Feichter, 1997; IPCC, 2001). However, the indirect effects are one of the most uncertain forcing mechanisms caused by anthropogenic aerosols.

In addition, the scattering and absorption of oncoming radiation (shortwave) by atmospheric aerosols are one of the major impacting mechanisms on the climate system. The direct effect of aerosols can modify the planetary albedo and reduce the amount of radiation reaching the terrestrial surface (IPCC, 2001). The radiative forcing due to anthropogenic aerosols may be equivalent in magnitude, but opposite in sign, to that of the greenhouse gases (IPCC, 1996). Whereas the atmospheric concentration of carbon dioxide (CO_2) and other infrared-active gases trap radiation and will result in warming of the Earth climate system, the direct effect of aerosols can exert a net cooling on the planet (Schwartz, 1996; IPCC 2001). Aerosols also absorb and emit terrestrial radiation (longwave) and, thus, produce a significant heating in the dense aerosol layer. The scattering and absorption by aerosols depend on their physical, chemical and optical characteristics, and are the primary causes of visibility impairment (Sloane, 1984). Unfortunately, the magnitude of this forcing still remains uncertain (IPCC 2001, Gomes and de Paula, 2003).

Due to their potential contributions to climate change, the study of aerosol radiative forcing is of great interest in the area of environmental sciences. In the 2001 IPCC report on its chapter five, Aerosol, their Direct and Indirect Effects, present a description of aerosol properties, their source and production mechanisms, and an analysis of the contributions of the uncertainties in the different factors needed to estimate both indirect and direct aerosol effects. For instance, on a global average, direct radiative forcing (DRF) from biomass burning aerosols range from -0.1 to -0.5 Wm^{-2} , where the main uncertainties are the single scattering albedo, the upscatter fraction, and the aerosol burden (IPCC, 2001). Hobbs et al. (1997) studied the radiative forcing from smoke aerosols during the biomass burning season in the Brazilian Amazon and found that the global mean direct forcing of the biomass burning aerosols was -0.3 Wm^{-2} . Grant et al. (1999) used a version of the NCAR CCM1 (National Center for Atmospheric Research -Community Climate Model) climate model to estimate the global average forcing of the sulfate and biomass burning aerosols. They found that the global average forcing of the biomass burning aerosols ranged from -0.23 to -0.16 Wm^{-2} .

In this work, it is presented a study of the mainly processes responsible for the direct effect of aerosols, in order to examine the regional impacts of the anthropogenic emission of the biomass burning aerosols. The goal of the proposed research is to link size distribution and optical properties of the biomass burning aerosols with the direct radiative forcing (DRF), in the context of regional climate change. In this paper, the focus was on biomass burning, which is the main source of carbonaceous aerosols (black carbon and organic carbon) over the tropics and the equatorial regions during dry season (Yamasoe et al., 2000). The sites selected were the Brazilian Amazon and Cerrado regions, which are regions of intense biomass burning activity. To accomplish this objective, it will be described the size distribution, chemical composition and optical properties of the biomass burning aerosols. The aerosols data were taken from the SCAR-B (Smoke, Clouds and Radiation-Brazil) experiment carried out in Brazil during 1995 (Kaufman et al., 1998).

The direct radiative forcing of the biomass burning aerosols (ΔF) was obtained from two simples radiative models. The direct radiative forcing at the top of atmosphere (TOA) was obtained from upward scattering model (USM) (Seinfeld and Pandis, 1998). The direct radiative forcing at the surface was obtained from downward scattering model (DSM) (Dimri and Jain, 1999). The radiative forcing within the atmosphere was derived from difference between of two radiative models. Finally, the results of the USM and DSM models are compared with the formulation proposed by Procópio et al. (2004).

2. Properties of atmospheric aerosols

Atmospheric aerosols (or particulate matter) and the surrounding gas, form complex three-phase system (solid, liquid, gaseous), which may transform to other compounds and are subjected to coagulation processes. Aerosols absorb and scatter light, with the efficiency of the processes being highly dependent on their size distribution, chemical composition and the wavelength of the incident radiation. The scattering efficiency is a function of particle size, morphology, and chemical composition, and is mainly due to particles in the accumulation mode ($0.1 \mu\text{m} < d_{ap} < 2.5 \mu\text{m}$). The light absorption by particles covers the whole radiation spectrum, and is largely due to the near-graphitic carbon, whose unique known sources are the incomplete combustion processes of fossil and biomass carbonaceous fuels (Castro et al., 1999).

Tropospheric aerosols have short atmospheric lifetimes, display a pronounced spatial variability and affect climate on a regional and global scale. Aerosols into the atmosphere arise from natural sources, such as volcanic dust, sea spray, wind-blown dust, biologic (pollen, bacteria, algae, leaf fragments, etc.) and anthropogenic sources, such as combustion of fossil fuel and biomass burning. Particulate matter can also be described as primary and secondary particles, based on their origin and formation process. Primary particles are emitted directly into the atmosphere by natural and anthropogenic sources, for instance, sea spray, mineral aerosol, smoke, soot, etc. Secondary particles are formed in the atmosphere by gas-to-particles conversion processes, for instance, sulfates, nitrates, and some organics.

2.1. Size distribution

Knowledge of the aerosol size distribution is of central importance in the study of the various aerosol behaviors in the atmosphere, such as atmospheric residence time, environmental effects (cloud and fog formation, degradation of visibility, etc). This physical property of the aerosols is also important for the identification of their chemical composition, origin, optical properties, and damage to human health (Pohojola et al., 2003). The variations in size distribution of the aerosols strongly influence the radiative properties, such as aerosol optical thickness ($\tau_{a\lambda}$), single scattering albedo ($\omega_{0\lambda}$), upscatter fraction (β) and asymmetry parameter (g_{λ}), (Masmoudi et al., 2003).

The size distribution of the atmospheric aerosols generally can be described as containing two modes as a function of the aerodynamic diameter of the particle (assuming a spherical shape): Fine mode with $d_a < 2.5 \mu\text{m}$ and coarse mode with $2.5 \mu\text{m} < d_a < 100 \mu\text{m}$. The fine mode is divided on the nuclei mode (also Aitken modes, $0.001 \mu\text{m} < d_a < 0.1 \mu\text{m}$) and accumulation mode ($0.1 \mu\text{m} < d_a < 2.5 \mu\text{m}$), Whitby and Cantrell (1976). The distinction between fine and coarse particles is essential because, in general, fine and coarse particles are originated separately, are removed from the atmosphere by different mechanisms, are transformed separately, and have different chemical and optical properties.

2.2. Chemical composition

The measurements of chemical composition of the airborne particles can be used to identify various properties of the aerosols, such as the various sources of the particles and their precursors, the optical properties, impact on the environment, effect on the air quality, and human health effects. It is also important to estimate the radiative effects of the aerosols. The chemical composition shows spatial and temporal variability and varies significantly with the particle size (Pandis et al., 1990). The chemical composition can be divided in two classes (Bizjak et al., 1999): (1) soil origin elements (Si, Al, Ca, Na, Fe and Ti) and are found in coarse mode particles; (2) enriched elements from natural and anthropogenic emissions, such as crustal material (Sc, Al); marine aerosol (Na, Cl); oil combustion (La, La/Sm, V), automobile traffic (Pb, Br, Zn), copper smelter (Cu), biomass burning (K); incinerator (Na, K, Cl, In, Hg); industrial urban areas (V, Zn, Se, Mo, Sb); Iron/Steel works (Fe, Zn, Se, Mo, Sb) and are found mainly in the fine size range.

2.3 – Optical properties

The optical properties of the atmospheric aerosols are based on specifications of the microphysical aerosols, such as size distribution and spectral refractive index under the assumption of homogeneous internally mixed spherical particles (Mie theory). However, aerosols in the atmosphere are generally not spherical in shape and this assumption is most critical for particles in the accumulation mode (Guyon et al. 2003). The aerosol optical properties show a temporal and spatial variability because of their atmospheric short lifetime (Masmoudi et al., 2003). Ristori et al., (2003) pointed out that the temporal variability is correlated with the different atmospheric scales process and the spatial variability is associated with local and regional events from natural and anthropogenic emissions, such as biomass burning and volcanic eruptions. The physical parameters involved in Mie calculations are the particle size (ρ_o) and the complex refractive index (m_r). The particle size is expressed as a dimensionless size parameter, $\rho_o = 2\pi r_o/\lambda$, where r_o is the mean radius of the particle and λ is the wavelength. The complex refractive index of the sphere relative to the surrounding medium is expressed as $m_r = n - ik$, where n is the real part with significant influence in the scattering properties of the aerosol and k is the imaginary part that characterizes its absorption.

3. Aerosol radiative properties

Aerosols and gases in the atmosphere scatter and absorb the solar radiation that enter in the Earth's atmosphere, and contribute to the light extinction. The reduction of sunlight intensity is called attenuation (or extinction). The rate of attenuation is expressed as light extinction coefficient (b_{ext}) and calculated by:

$$b_{ext} = b_{sp} + b_{ap} + b_{sg} + b_{ag} \quad (1)$$

where b_{sp} and b_{ap} refer to components due to scattering and absorbing of light by particles and b_{sg} and b_{ag} refer to components due to scattering and absorbing by gases. The extinction of a beam of parallel monochromatic radiation through the atmosphere can be calculated by the Beer-Lambert law. It is the basis for all the calculations of the spectral optical thickness for each wavelength λ (Iqbal, 1983):

$$I(\lambda) = I_o(\lambda) \exp(-\tau_{T\lambda} m) \quad (2)$$

where $I(\lambda)$ is the spectrometer signal; $I_o(\lambda)$ is the extraterrestrial spectrometer signal, m is the relative optical air mass (or optical path length) and $\tau_{T\lambda}$ is the total spectral optical thickness. $\tau_{T\lambda}$ of the atmosphere corresponds to the integrated value of the extinction coefficient at each altitude by atmospheric thickness. The optical thickness indicates the magnitude of scattering and absorption of the solar radiation. The total spectral optical thickness includes the light attenuation by scattering (air molecules and aerosols) and by absorption (gases and aerosols). The aerosol optical thickness ($\tau_{p\lambda}$) may be obtained by eliminating from $\tau_{T\lambda}$ the contributions due Rayleigh dispersion and the absorption by the atmospheric components:

$$\tau_{a\lambda} = \tau_{T\lambda} - \tau_{R\lambda} - \tau_{O_2\lambda} - \tau_{O_3\lambda} - \tau_{H_2O\lambda} - \tau_{NO_2\lambda} \quad (3)$$

where $\tau_{R\lambda}$ is the optical thickness of atmosphere due Rayleigh scattering; $\tau_{O_2\lambda}$ is the optical thickness of oxygen; $\tau_{O_3\lambda}$ is the optical thickness of ozone; $\tau_{H_2O\lambda}$ is the optical thickness of water vapour caused by the absorption bands near; and $\tau_{NO_2\lambda}$ is the optical thickness of nitrogen dioxide. The single scattering albedo ($\omega_{o\lambda}$) is one parameter generally used for characterizing the total reflectance of the atmosphere. It is a measure of the relative importance of scattering and absorption by the particles. When is considered individual particles, $\omega_{o\lambda}$ is calculated by ratio between scattering efficiency and extinction efficiency:

$$\omega_{\delta\lambda} = \frac{b_{ep}}{b_{extp}} = \frac{b_{ep}}{b_{ep} + b_{ap}} \quad (4)$$

The single scattering albedo is dependent of characteristics of the particles, such as the wavelength, the way how the scattered field is measured, refractive index, chemical composition and size distribution. The angular distribution of the scattered radiation at a given wavelength is the aerosol phase function ($P_{a\lambda}$). The most common approximation to the phase function is the two-term Henyey-Greenstein function. This formulation is used in modelling of the energy radiative transfer in region that may be affected by different kinds of aerosols, depending on the atmospheric synoptic conditions. The two-term Henyey-Greenstein function, modified by Irvine (1965) can be express as:

$$P_{a\lambda} = \frac{w(1-g_1^2)}{(1+g_1^2-2g_1\cos\Theta)^{0.5}} + \frac{(1-w)(1-g_2^2)}{(1+g_2^2-2g_2\cos\Theta)^{1.5}} \quad (5)$$

where Θ is the scattering angle; w , g_1 and g_2 are asymmetry parameters. The asymmetry parameter is to express the distribution of scattered light. The microphysics of the aerosols influences the radiative forcing through the upscatter fraction (β), which exhibits a dependence on particle size (ρ_o) and solar zenith angle (θ_o) as a consequence of the particle size of the phase function, i.e., the angular distribution of scattered radiation (Schwartz, 1995). It is defined as the fraction of solar radiation that is scattered by particle into the upward hemisphere that escapes to space. The upscatter fraction as a function of ρ_o and θ_o was developed by Wiscombe and Grams (1976). Figure (1) gives β as a function of cosine of solar zenith angle $\mu_o = \cos \theta_o$, and particle radius at $\lambda = 0.55 \mu\text{m}$.

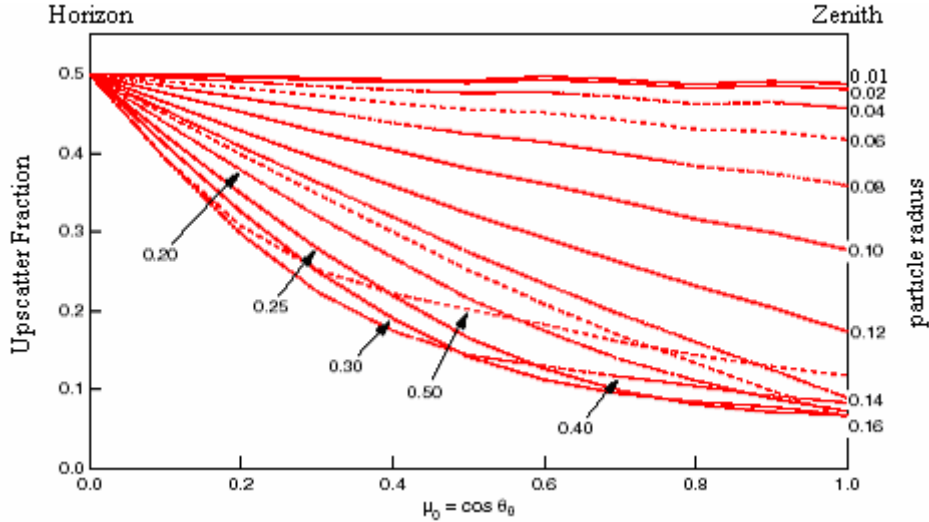


Figure 1. Upscatter fraction as function of the cosine of the solar zenith angle for indicated values of particle radius and $\lambda = 550 \text{ nm}$. Source: Schwartz (1996).

A parameter that is derived from the phase function is the asymmetry parameter (g_λ), which represents the weighted average of the β . In the present study, in order to estimate the asymmetry parameter, it was applied the following formulation proposed by Sokolik and Golitsyn (1993):

$$g_\lambda = -B_1\rho_o^2 + B_2\rho_o + B_3 \quad (6)$$

where ρ_o is the size of particle; and B_i are coefficients that depend only of imaginary part, k , of the complex refractive index of aerosols m_r . The coefficients B_i are expressed as:

$$B_1 = 0.0007 + 0.0281k ; \quad B_2 = 0.0217 + 0.8287k ; \quad B_3 = 0.6243 + 2.1559k \quad (7)$$

4. Direct radiative forcing of the atmospheric aerosols

The direct radiative forcing (DRF) of the aerosols is defined as the ratio of the change in the surface reaching solar radiation intensity, ΔF^\downarrow in W/m^2 or the upscattered solar radiation flux ΔF^\uparrow , leaving the atmosphere to the change in the column aerosol optical depth, $\Delta\tau_{a\lambda}$. The forcing efficiencies at the surface, $\Delta F^\downarrow/\Delta\tau_{a\lambda}$, and at the top of the atmosphere (TOA), $\Delta F^\uparrow/\Delta\tau_{a\lambda}$, are estimated separately and the difference between the two terms is defined as the net aerosol

absorption efficiency within the atmosphere. The net-flux changes in the radiation budget can modify the surface temperature and thus the sensible and latent heat fluxes into the lowest layers of the atmosphere (Yu et al., 2003). This process causes alterations in the temperature vertical profile of the atmosphere, height of the boundary layer, affecting the atmospheric stratification, regional atmospheric circulation, cloud formation, precipitation and energy conversion (e.g. surface heating, convection, evaporation and thermal emission) (Procópio et al., 2004). The magnitude of the aerosol radiative forcing at any location and time depends on the amount of the aerosols, their optical properties, underlying surface albedo and the solar zenith angle that describe the interaction of aerosols with the solar radiation (Penner et al., 1994). In the present study, it was used two simple radiative models, in order to analyze the effect of the radiative forcing from aerosol layer underlying the Earth's surface. These models were used to determine the direct radiative forcing (DRF) at the top of atmosphere (TOA), at the surface. The difference between the two models was used to determine the radiative forcing at the atmosphere.

4.1.1. Upward scattering model of an aerosol layer for radiative forcing at the TOA

The upward scattering model (USM) was proposed by Seinfeld and Pandis (1998). The direct radiative forcing is estimated assuming an optically thin layer of aerosol where the multiple reflections within of the aerosol layer are neglected. Consider an aerosol layer, where a direct solar beam impinge on the layer with the assumption that the solar zenith angle is $\theta_0 = 0^\circ$, Fig. (2).

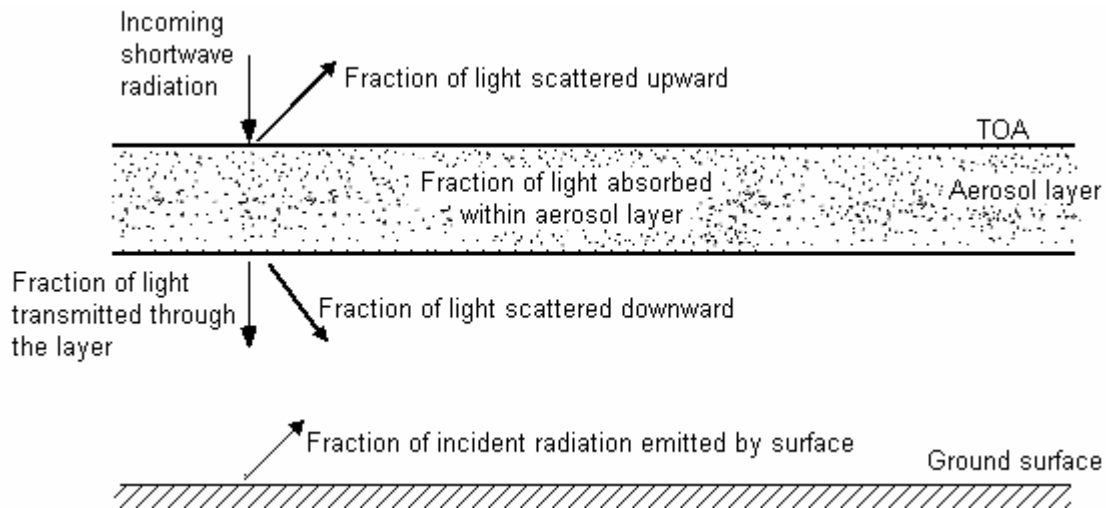


Figure 2. Upward scattering model of an aerosol layer above the Earth's surface (Seinfeld and Pandis, 1998).

In Fig. (2) the solar radiation flux incident (F_o) on the aerosol layer underlying the Earth's is decomposed into following fractions:

- (1) - Fraction of light scattered upward: $r = (1 - e^{-\tau_{a\lambda}}) \omega_{o\lambda} \beta_\lambda$
- (2) - Fraction of light scattered downward: $s = \omega_{o\lambda} (1 - \beta_\lambda) (1 - e^{-\tau_{a\lambda}})$
- (3) - Fraction of light absorbed within aerosol layer: $A = (1 - \omega_{o\lambda}) (1 - e^{-\tau_{a\lambda}})$
- (4) - Fraction of the incident beam transmitted through the layer: $T_r = \exp(-\tau_{a\lambda})$
- (5) - Total fraction of radiation incident on the layer transmitted downward: $t = e^{-\tau_{a\lambda}} + \omega_{o\lambda} (1 - \beta_\lambda) (1 - e^{-\tau_{a\lambda}})$.

In the Fig. (2) the total fraction of the radiation incident on the surface is t and the fraction of the radiation incident emitted from the surface is $R_s t$, where R_s is the mean albedo of the underlying Earth's surface. The radiation emitted from the surface ($R_s t$) interacts with the aerosol layer and some is backscattered back to the Earth ($r R_s t$), some is absorbed by aerosol layer ($A R_s t$) and some is scattered upward ($R_s t^2$). The reflected fraction from the aerosol layer ($r R_s t$), interacts with surface and to produces a the reflected fraction ($r R_s^2 t$), which interacts with the aerosol layer to produce a reflected fraction toward the surface ($r^2 R_s^2 t$), an absorbed fraction by aerosol layer ($A r R_s^2 t$) and scattered upward fraction ($R_s^2 t^2$). The fraction reflected downward ($r^2 R_s^2 t$), is itself reflected off the Earth's surface to produce the fraction ($r^2 R_s^3 t$), which interacts with aerosol layer to produces higher order of the upward transmitted fraction. The total upward flux of an aerosol layer underlying an atmospheric layer is given by

$$F_{TOA} = r F_o^\downarrow + R_s F_o^\downarrow t^2 + r R_s^2 t^2 F_o^\downarrow + r^2 R_s^3 t^2 F_o^\downarrow + \dots \quad (8.a)$$

$$F_{TOA} = F_o^\downarrow [r + R_s t^2 (1 + r R_s + r^2 R_s^2 + r^3 R_s^3 + \dots)] \quad (8.b)$$

where F_o^\downarrow is the downward solar flux incident top of the atmosphere. Since R_s and r are minor that 1, the series in the small parentheses is: $1 + r R_s + r^2 R_s^2 + r^3 R_s^3 + \dots = 1 / (1 - R_s r)$. Therefore, the total upward reflected flux can be written as:

$$F_{TOA} = \left[r + \frac{t^2 R_s}{1 - R_s r} \right] F_o^\downarrow \quad (9)$$

In the atmosphere without aerosols: $\tau_{a\lambda} = 0$, $t=1$ and $r = 0$, thus the total upward reflected flux from surface is $F_r^\uparrow = R_s F_o$. The aerosol direct radiative forcing at the TOA is given by difference between the total upward reflected flux with and without aerosols:

$$\Delta F_{TOA} = F_o^\downarrow \left[r + \frac{t^2 R_s}{1 - R_s r} \right] - F_o^\downarrow R_s \quad (10)$$

where ΔF_{TOA} is the change in forcing at the TOA. In the Eq. (10) is assumed that only the aerosols in the atmosphere produce scattering and absorption. However, the radiation transmission through the atmosphere is also affected by absorption and scattering of the atmospheric molecules. Therefore, the incident flux F_o is $T_a F_o$, where T_a is the fractional transmittance of the atmosphere, i.e., the fraction of light transmitted by the atmospheric layer above the aerosol layer. Therefore, the DRF of the aerosols at the TOA, as result of the change in albedo occurring in the cloud free area in the atmosphere is given by (Seinfeld and Pandis, 1998):

$$\Delta F_{TOA} = -F_o^\downarrow (1 - A_c) T_a^2 \left[\left(r + \frac{t^2 R_s}{1 - R_s r} \right) - R_s \right] \quad (11)$$

where the minus sign denotes the convention that a negative forcing is a cooling influence in the climate change.

4.1.2. Downward scattering model of an aerosol layer for radiative forcing at the surface

The downward scattering model (DSM) was proposed by Dimri and Jain (1999) to the estimate radiative forcing at the Earth's surface. However, due the high values of DRF of the aerosols that were obtained, in the present study, it was necessary included in the model, the fractional atmospheric transmittance, T_a . Figure (3) shows an aerosol layer underlying the Earth's surface, where multiple reflections within of aerosol layer are considered.

In this model aerosols are assumed to be monodisperse, i.e., same size distribution, same chemical composition, and homogeneously distributed within of the layer. Consider that the infrared radiation (F_s^\uparrow) emitted from Earth's surface interacts with the aerosol layer, and some is backscattered back to the Earth ($R_e F_s^\uparrow$), some is absorbed within of aerosol layer ($A R_e F_s^\uparrow$), and some is transmitted toward space by aerosol layer ($T_r R_e F_s^\uparrow$), where R_e , A and T_r are reflectance, absorptance and transmittance coefficients of the aerosol layer, respectively. The reflected component back downward to the surface from the aerosol layer ($R_e F_s^\uparrow$), is itself is reflected off the surface and to produces a reflected component from the surface ($\alpha_s R_e F_s^\uparrow$), where α_s is the absorptivity of the ground. This reflected component ($\alpha_s R_e F_s^\uparrow$), from surface interacts with the aerosol layer and to produce a reflected component ($\alpha_s R_e^2 F_s^\uparrow$) toward the ground, an absorbed component ($A \alpha_s R_e F_s^\uparrow$), and transmitted component toward to space ($T_r \alpha_s R_e F_s^\uparrow$) and so, successively. The first component absorbed by aerosol layer ($A R_e F_s^\uparrow$), cause heat within layer and emits a flux ($\varepsilon A R_e F_s^\uparrow$) toward the Earth's surface, where ε is the emissivity of the aerosol layer. This emitted flux ($\varepsilon A R_e F_s^\uparrow$) is reflected from the Earth's surface and to produce the reflected component ($\alpha_s \varepsilon A R_e F_s^\uparrow$), which interacts with aerosol layer to produces higher order reflected and emitted fluxes. The total flux directed toward the Earth's surface to produce the net flux ΔF_s^\uparrow , which represent the aerosol direct radiative forcing at the surface is given by

$$\begin{aligned} \Delta F_{SUP} = & [R_e F_s^\uparrow + \alpha_s R_e^2 F_s^\uparrow + \alpha_s^2 R_e^3 F_s^\uparrow + \dots] + [\varepsilon A F_s^\uparrow + \varepsilon A \alpha_s R_e F_s^\uparrow + \varepsilon A \alpha_s^2 R_e^2 F_s^\uparrow + \dots] + \\ & [\varepsilon \alpha_s R_e A F_s^\uparrow + \varepsilon A \alpha_s^2 R_e^2 F_s^\uparrow + \varepsilon A \alpha_s^3 R_e^3 F_s^\uparrow + \dots] + [\varepsilon^2 A^2 \alpha_s F_s^\uparrow + \varepsilon^2 A^2 \alpha_s^2 R_e F_s^\uparrow + \varepsilon^2 A^2 \alpha_s^3 R_e^2 F_s^\uparrow + \dots] + \dots \end{aligned} \quad (12)$$

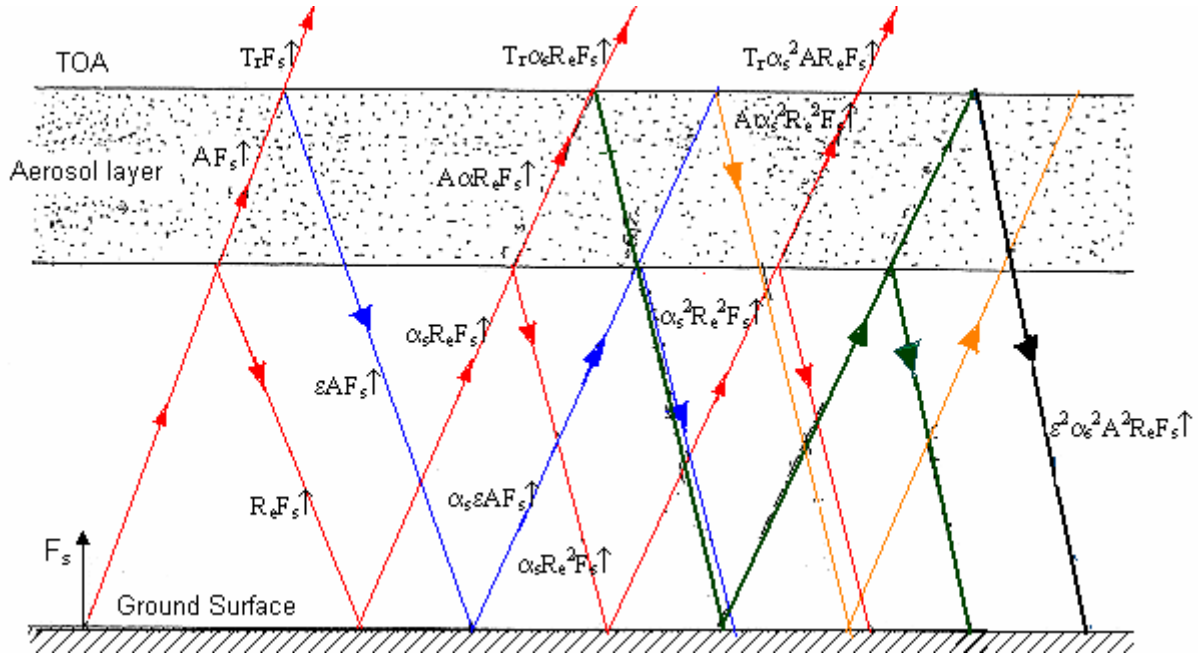


Figure 3. Downward scattering model of an aerosol layer above the Earth's surface (Dimri and Jain, 1999)

The total flux on the surface due an aerosol layer underlying an atmospheric layer can be expressed by following equation,

$$\Delta F_{SUP} = F_s \uparrow [1 + \epsilon A \alpha_s] [\epsilon A + R_e] [1 + \alpha_s R_e + \alpha_s^2 R_e^2 + \alpha_s^3 R_e^3 + \dots] \quad (13)$$

Since R_e and α_s are < 1 , the series in the last brackets is: $1 + \alpha_s R_e + \alpha_s^2 R_e^2 + \dots = 1 / (1 - \alpha_s R_e)$ and the product $\epsilon A \alpha_s$ is very small. Therefore, the total upward reflected flux reduces to

$$\Delta F_{SUP} = F_s \uparrow (\epsilon A + R_e) \left(\frac{1}{1 - \alpha_s R_e} \right) \quad (14)$$

In the Eq. (14), it is assumed that only the aerosol in the atmosphere produces scattering and absorption. However, the radiation transmission through the atmosphere is also affected by absorption and scattering of the atmospheric molecules. Therefore, in the present study, it was included the atmospheric transmittance T_a in the model proposed by Dimri and Jain, (1999). Therefore, the aerosol direct radiative forcing at surface that is defined as the aerosol-induced change in the infrared radiation emitted from the surface (ΔF_{sup}), as a function of wavelength is given by

$$\Delta F_{SUP} = F_s \uparrow (\epsilon_{a\lambda} A + R_e) T_a^2 \left(\frac{1}{1 - \alpha_{s\lambda} R_e} \right) \quad (15)$$

where $F_s \uparrow = \sigma T_{sup}^4$, T_{sup} is the surface temperature and σ is the Stefan Boltzmann constant ($\sigma = 5.67 \times 10^{-8} \text{ W m}^{-2} \text{ T}^{-4}$).

The reflectance R_e and the absorptance A of the aerosol layer can be expressed as $R_e = \omega_{o\lambda} (1 - g_\lambda) (1 - e^{-\tau_{a\lambda}})$ and $A = (1 - \omega_{o\lambda}) (1 - e^{-\tau_{a\lambda}})$, respectively, where $\tau_{a\lambda}$ is the aerosol optical thickness, $\omega_{o\lambda}$ is the single scattering albedo, and g_λ is the asymmetry parameter.

5. Data sets and site of study

Forcing $\Delta F \uparrow$ was evaluated for the values of the optical properties (aerosol optical thickness and single scattering albedo) and size distribution of the biomass burning aerosols from *in situ* measurements obtained from literature. These data sets were retrieved from ground based-measurements and satellite-measurements over dark surfaces from field campaigns on biomass burning carried out in the Brazilian Amazon and Cerrado regions. The field campaigns were carried out during the dry season from SCAR-B (Smoke, Clouds and Radiation-Brazil) experiment (August-September 1995), which comprises an experiment to study the biomass burning, emphasized measurements of the surface biomass,

fires, smoke aerosol and radiation (Kaufman et al., 1998). The smoke aerosols were assumed as a solid, spherical core of black carbon surrounded by viscous organic liquids because the combustion particles were considered internally mixed. The total column smoke ($\tau_{a\lambda}$), at $\lambda = 0.55 \mu\text{m}$ was estimated using the observed radiance from ground-based sun-photometer from AERONET (AEROSOL ROBOTIC NETWORK). This monitoring program is a federation of ground-based remote sensing aerosol network conducted by NASA (<http://aeronet.gsfc.nasa.gov>). The $\omega_{0\lambda}$ and solar zenith angle were retrieved from the visible channel ($\lambda = 0.63 \mu\text{m}$) of the Advanced Very High Resolution Radiometer (AVHRR) sensor on board the National Oceanic and Atmospheric Administration (NOAA-14) polar orbiting satellite. Table (1) shows the locations, latitude, longitude, International Geosphere Biosphere Programme (IGBP) surface types, aerosol optical thickness and single scattering albedo for a total of 10 sites selected where the sun-photometers were located during SCAR-B experiment (Li et al., 1999). The results of Tab. (1) showed larger differences in the spatial distribution of the aerosol optical thickness (AOT) and was found to range 0.59 to 2.25. The single scattering albedo (SSA) showed smaller differences in the spatial distribution of in the region of study, which ranged to 0.753 to 0.906.

Table 1. The locations, their latitude and longitude, IGBP surface ecosystems, means and standard deviations of AOT and SSA. Source: Li et al., 1999.

Site	Location (Latitude, Longitude)	IGPB type	AOT (0.55 μm)		SSA (0.55 μm)	
			$\mu_{\tau_{a\lambda}}$	$\sigma_{\tau_{a\lambda}}$	$\mu_{\tau_{a\lambda}}$	$\sigma_{\omega_{0\lambda}}$
Alta Floresta	(-9.92, -56.00)	Broadleaf Forest	2.25	0.75	0.862	0.044
Ariquims	(-9.89, -63.00)	Broadleaf Forest	2.45	NA	0.886	NA
Campo Verde	(-15.55, -55.16)	Savanna	1.71	NA	0.753	NA
Cuiabá	(-15.50, -56.00)	Savanna	0.92	0.44	0.831	0.053
El Refugio	(-14.75, -62.03)	Savanna	1.68	0.60	0.849	0.069
Ji Paraná	(-10.85, -61.79)	Savanna	2.59	0.60	0.906	0.020
Pantanal	(-16.38, -56.62)	Wetland	1.54	0.22	0.859	0.051
Potosi Mine	(-9.27, -62.86)	Broadleaf Forest	2.50	0.71	0.891	0.031
Tukurui	(-3.71, -49.67)	Broadleaf Forest	0.95	NA	0.835	NA
Uberlândia	(-18.89, -48.27)	Grassland	0.59	0.19	0.873	0.055

6. Results and discussion

In order to estimate the radiative forcing at the TOA under cloud free conditions in the USM model, it was necessary to obtain in the literature the values of the upscatter fraction, mean surface albedo, atmospheric transmittance and incident solar flux. In this case β was obtained as a function of the mean radius of the particle r_o , and solar zenith angle θ_o . The size distribution of the smoke aerosols was obtained by Ross et al. (1998) during the SCAR-B experiment. This size distribution was assumed to be lognormal with a mode radius of $0.14 \mu\text{m}$, and a standard deviation of 1.35. During the SCAR-B experiment the solar zenith angle ranged between $0-70^\circ$ and the relative azimuth angle ranged from $0-180^\circ$, in 10° intervals. The solar zenith angle was 30° and the relative azimuth between the sun and the sensor was 40° (Li et al., 1999). Therefore, the upscatter fraction was obtained from Fig. (1), where β is as a function of the solar zenith angle ($\mu_o = \cos \theta_o$) and particle radius, for $\lambda = 0.55 \mu\text{m}$. In the present study β was equal to 0.15 with $r_o = 0.14 \mu\text{m}$, $\theta_o = 30^\circ$ and $\mu_o = 0.86$. The surface was assumed to be Lambertian with a surface albedo of 0.10 ($\lambda = 580$ to $680 \mu\text{m}$) (Li et al., 1999). The mean values of atmospheric transmittance above the aerosol layer, $T_a = 0.76$ was obtained from Schwartz (1996) and Seinfeld and Pandis (1998), and the incident solar flux $F_o = 343 \text{ Wm}^{-2}$, from Seinfeld and Pandis (1998).

In the DSM model to assess the radiative forcing on the surface, it was necessary to obtain the values of the asymmetry parameter, emissivity of the aerosol layer, infrared radiation emitted from surface, absorptivity and temperature of the Earth surface. The asymmetry parameter at $\lambda = 0.55$ was obtained using Eq. (2). In this case, it was considered the size parameter ρ_o from Eq. (1) and coefficients B_i , from Eq. (8). These coefficients are depend only on the of imaginary part k , of the complex refractive index of the aerosols. In the present study, the imaginary part of the complex index of refraction of the biomass burning aerosols was $k = 0.013$, obtained from two field campaigns at a remote rainforest site in Amazon Basin (Guyon et al. 2003). Therefore, the value of the asymmetry parameter adopted was $g_\lambda = 0.7015$. The emissivity of aerosol layer $\varepsilon_{a\lambda} = 0.0418$ was obtained from experiments carried out by Charalampopoulos and Chang (1991). These authors investigated various properties of soot-particle agglomeration for a premixed flame and the spectral emissivity was obtained for the wavelength range 0.4 to $6.7 \mu\text{m}$. This emissivity is considered in the present study because biomass burning is the major source of the soot (black carbon or carbon graphitic) (Simoneit 2002). The absorptivity of the surface, $\alpha_{s,\lambda} = 0.97$ was obtained from a study performed by (Petitcolin and Vermote, 2002). These authors retrieved surface emissivity from Moderate Resolution Imaging Spectroradiometer (MODIS) data on-board the EOS-TERRA satellite. The value of surface temperature considered was

T = 288 K. This parameter is considered in several studies (Seinfeld and Pandis, 1998). Therefore, $F_s^\uparrow = 5.67 \times 10^{-8} \times 288^4 = 390 \text{ Wm}^{-2}$. Table (2), summarizes the parameters that were used in both USM and DSM models to estimate the aerosol direct radiative forcing at the TOA and at the surface.

Table 2. Parameters used in the modelling of direct radiative forcing from biomass burning aerosol.

Parameters	Values	Reference
r_o	0.14 μm	Ross et al. (1998)
θ_o	30°	Li et al. (1999)
β_λ	0.15	Figure 1 (Schwartz, 1996)
T_a	0.76	Schwartz (1996); Seinfeld and Pandis (1998)
F_o	343 Wm^{-2}	Seinfeld and Pandis (1998)
R_s	0.10	Liu et al. (1999)
K	0.013	Guyon et al. (2003)
g_λ	0.7015	Equation 7 (Sokolik and Golitsyn, 1993)
$\varepsilon_{a\lambda}$	0.0418	Charalampopoulos and Chang (1991)
$\alpha_{s,\lambda}$	0.97	Pettilcolin and Vermote (2002)
T_s	288 K	Seinfeld and Pandis (1998)
F_s^\uparrow	390 Wm^{-2}	Stefan-Boltzmann Law

6.1. Model validation

In order to assess the DRF of the biomass burning aerosols at the TOA and at the surface Eq. (12) and (16) were applied, respectively. The DRF in the atmosphere was obtained by subtracting the DRF at the TOA from the DRF at the surface. To validate the modelling, it was carried a comparison between the results of DRF of the biomass burning aerosol with the formulation proposed by Procópio et al. (2004). These authors performed an analysis of seven years of the clear-sky aerosol direct radiative forcing for two locations in the Amazon Region, heavily impacted by biomass burning emissions during the dry season. Table (3) shows the formulation that was applied by Procopio et al. (2004) to assess DRF of the biomass burning aerosols, at $\lambda = 0.55 \mu\text{m}$. Table (4) shows the results of DRF at $\lambda = 0.55 \mu\text{m}$ from biomass burning aerosols during SCAR-B experiment carried out in 10 sites of the Brazilian Amazon and cerrado regions during dry season (August-September, 1995). In Tab. (4) column A are results of the models applied in the present work and column B are results from application of the formulation proposed by Procópio and co-workers.

Table 3. Regression coefficients for the aerosol radiative forcing at the surface and TOA. Source: Procópio et al. (2004).

^(a) $ARF_{24h} = a(\tau_{a\lambda})^3 + b(\tau_{a\lambda})^2 + c\tau_{a\lambda} + d$					
	<i>a</i>	<i>b</i>	<i>C</i>	<i>d</i>	<i>R</i> ²
Surface	0	5.04	-51.6	3.92	0.999
TOA	-0.95	6.71	-16.5	1.57	0.994

^(a)ARF_{24h} is the 24 h aerosol radiative forcing (Wm^{-2}), $\tau_{a\lambda}$ is the daily average AOT (at $\lambda = 0.55\mu\text{m}$) and *a*, *b*, *c* and *d* are regression coefficients. Table (4) shows the results of DRF of the biomass burning aerosol in 10 sites at Brazilian Amazon and cerrado region during the dry season (August-September, 1995).

Table 4. The DRF of the biomass burning aerosols at the top of atmosphere (TOA), at the surface and at the atmosphere.

Site	DIRECT RADIATIVE FORCING (Wm^{-2})					
	TOA (0.55 μm)		Surface (0.55 μm)		Atmosphere (0.55 μm)	
	A ⁽¹⁾	B ⁽²⁾	A ⁽¹⁾	B ⁽²⁾	A ⁽¹⁾	B ⁽²⁾
Alta Floresta	- 14.71	-12.41	- 68.25	- 86.66	53.54	74.25
Ariquims	- 16.27	-12.55	- 72.38	- 92.25	56.11	79.70
Campo Verde	- 8.46	- 11.77	- 52.81	- 69.58	44.35	57.81
Cuiabá	- 8.57	- 8.67	- 40.41	- 39.29	31.84	30.62
El Refugio	- 12.69	- 11.72	- 59.51	- 68.54	46.82	56.83
Ji Paraná	- 17.53	- 12.66	- 75.48	- 95.91	57.95	83.25
Pantanal	- 12.67	-11.39	- 57.69	- 63.59	45.02	52.20
Potosi Mine	- 16.61	-12.59	- 73.29	- 93.58	56.68	80.99
Tukurui	- 8.89	- 8.86	- 41.54	- 40.55	32.65	31.69
Uberlândia	- 7.36	- 6.02	- 30.08	- 24.77	22.72	18.75

(1) Results of the present study and (2) results of the formulation proposed by Procópio and co-workers.

The values of the DRF at the top of atmosphere obtained from upward scattering model (USM) (Seinfeld and Pandis, 1998) tends to overestimate the values of the DRF when compared to the formulation proposed by Procópio et al. (2004) for values of AOT ($\lambda = 0.55\mu\text{m}$) greater than 1.0. However, occurred a discrepancy for $\tau_{a\lambda} = 1.71$ (Campo Verde site). When AOT ($\lambda = 0.55\mu\text{m}$) is smaller than 1.0 the values of both methods were approximately equals, except when $\tau_{a\lambda} = 0.59$ (Uberlândia site). In general, regional DRF from biomass burning aerosol at the TOA from USM model showed greater variation spatial, ranged from -7.36 to -17.53 Wm^{-2} in the region of study. The results suggested that the magnitude of the forcing tend decreases with decreasing aerosol optical thickness. The results of the both methodologies indicated that DRF of the biomass burning aerosols could induce a cooling at the TOA.

The value of the DRF at the surface obtained from downward scattering model (DSM) (Dimri and Jain, 1999) tends to sub-estimate the values of the DRF when compared to the formulation by Procópio and co-workers. When AOT ($\lambda = 0.55\mu\text{m}$) was about 0.9 the values of direct radiative forcing from DSM and formulation proposed by Procópio et al. (2004) were approximately equal. Only when AOT ($\lambda = 0.55\mu\text{m}$) was 0.59 (Uberlândia site) the value of the DSM model was greater than formulation of the Procópio and co-workers. In general, regional DRF from biomass burning aerosols at the surface from DSM model also showed greater variation spatial range from -30.08 to -75.48 Wm^{-2} . The results also suggested that the magnitude of the forcing tends decreases with the decreasing of aerosol optical thickness. The results of the both methodologies indicated that DRF of the biomass burning aerosols could induce a cooling at the surface.

The values of the DRF from biomass burning aerosols at the atmosphere, it was obtained by subtracting ΔF_{TOA} from ΔF_{sup} . The results showed that, regionally, aerosols can cause significant warming due absorption of the solar radiation. This possible warming can be caused mainly by black carbon aerosols emitted into the atmosphere as a byproduct of biomass burning. The black carbon aerosol may be considered as the only component of the aerosol phase which strongly absorbs the solar radiation in the visible and near-infrared part of the spectrum, where most of the solar energy is distributed (Latha and Badarinath, 2003).

7. Conclusion

In this study two radiative models were applied at the TOA and at the surface in order to estimate the direct effect of aerosol from biomass burning in the Brazilian Amazon and Cerrado regions during the dry season. The direct radiative forcing was obtained using aerosol optical thickness and single scattering data and other aerosol properties, namely the upscatter fraction, asymmetry parameter and particle size, collected in the literature. Regional distribution of aerosols properties was computed by using data from SCAR-B experiment (Kaufaman et al., 1998), carried out during August-September 1995 (Li et al., 1999). Also it was presented results of DRF from the formulation proposed by Procópio et al. (2004) in order to it compare with the results of the two models applied in the present study.

At the TOA the comparison reveals that upward scattering model (Seinfeld and Pandis, 1998) tends to overestimate the values of DRF when compared to the formulation proposed by Procópio and co-workers for AOT ($\lambda = 0.55\mu\text{m}$) greater than 1.0. At the surface the comparison reveals that downward scattering model (Dimri and Jain, 1999) tends to sub-estimate the values of DRF when compared to the formulation proposed by from Procópio and co-workers. The results of the application of USM and DSM showed that the biomass burning aerosols could cause cooling at the TOA and at the surface, respectively, due to the negative sign obtained in the direct radiative forcing. The values of the DRF from biomass burning aerosols at the atmosphere showed that, regionally, aerosols can cause significant warming due to the absorption of solar radiation. The comparison of USM at the TOA and of DSM at the surface with the formulation proposed by Procópio and co-workers suggested that the magnitude of aerosol forcing is still uncertain. Theses uncertainties may be related with uncertainties in the formulation of both models and uncertainties in the measurements of the aerosol optical properties. On the other hand, the upward scattering model and downward scattering model were capable to estimate DRF within the uncertainty range obtained by formulation proposed by Procópio et al. (2004).

The results from the application of simple radiative models support the general knowledge that the biomass burning in Amazon Basin may cause adverse effects on the local/regional climate system. The possible direct effects of aerosols on the climate change are related with cooling at the top of atmosphere and at the Earth's surface and warming at the atmosphere. Despite the larger number of research which investigated the influence of biomass burning aerosol in the Amazonian climate system, continued research is needed to understand better the possible direct effects of the aerosols on the local/regional climate.

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