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Effect of absorbing aerosols on global radiation budget

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Abstract. An expression for a globally averaged value of direct radiative forcing by absorbing aerosols is derived and applied to the case of smokes produced by biomass burning. It is shown that the direct radiative forcing due to the biomass burning aerosols is a sensitive function of the size distribution of aerosol particles. For the range of measured size distributions of smoke aerosols the direct radiative forcing varies between -0.2 and $-1.1 W/m^2$.

Introduction

Aerosols reflect a part of incoming solar radiation back to space (direct effect) and they modify cloud droplet size distribution and perhaps cloud life cycle (indirect effect). Charlson et al. (1992) estimate the direct globally averaged radiative forcing due to anthropogenic sulfate aerosol to be $-1 W/m^2$ with additional indirect effect by modifying cloud albedo of around $-1 W/m^2$. Penner et al. (1992) suggest that an additional globally averaged direct radiative forcing of about $-1 W/m^2$ is due to radiative effects of smokes produced by biomass burning. Another $-1 W/m^2$ in radiative forcing is attributed to indirect effects of anthropogenic smokes [*Penner et al.* , 1992]. Penner et al. (1992) note that the estimated radiative effects of anthropogenic sulfate and smoke aerosols had been probably exaggerated because otherwise a decrease of the global temperature should have been observed since the last century.

The direct radiative effect of anthropogenic sulfate aerosol have been investigated recently by Kiehl and Briegleb (1993) and Taylor and Penner (1994) using three dimensional models. The estimated direct globally averaged radiative forcing varied from $-0.3 W/m^2$ [*Kiehl and Briegleb*, 1992] to $-0.9 W/m^2$ [*Taylor and Penner*, 1994]. The indirect aerosol radiative forcing is even more complicated to estimate due to the current lack of understanding of the cloud-aerosol interaction.

In this note we consider the direct effect of anthropogenic smoke aerosol. Specific features of our investigation are (a) derivation of the change of planetary albedo due to an aerosol layer that includes absorbing properties of aerosols and (b) Mie scattering calculation of the smoke particle upscattering fraction that shows a strong dependence on the aerosol size distribution.

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Aerosol Radiative Forcing

The globally averaged direct aerosol radiative forcing, ΔF_R, was calculated by Charlson et al. (1992) using the expression

$$\Delta F_R = -\frac{S_0}{4} T_{atm}^2 (1-N)(1-a)^2 2\beta\tau_{sc} \qquad (1)$$

where S_0 is a solar constant, T_{atm} is the transmittance of the atmosphere above the aerosol layer, N is the fraction of sky covered by clouds, a is the albedo of underlying surface, β is the fraction of radiation scattered by aerosol into the upper hemisphere and τ_{sc} is the aerosol layer scattering optical thickness. The above expression gives the radiative forcing due to the change of reflectance of the earth-aerosol system. It can be derived from the radiative transfer equation for the case of optically thin aerosol layer ($\tau_{sc} \ll 1$) composed of nonabsorbing aerosol particles.

The physical meaning of individual terms in eq. (1) is the following; $S_0/4$ is the globally averaged incident solar flux at the top of the atmosphere, T_{atm}^2 reduces the incident flux and the reflected flux by the transmittance of the atmosphere above the aerosol layer, $1-N$ comes from the assumption that albedo changes due to aerosol is significant only in the cloud free atmosphere.

The last term in eq. (1)

$$\Delta R = (1-a)^2 2\beta\tau_{sc} \qquad (2)$$

represents the albedo increase due to a nonabsorbing aerosol layer.

The radiative transfer equation for an optically thin atmosphere leads to the following expressions for the planar albedo $R(\mu_0)$ and transmission $T(\mu_0)$ of an isolated optically thin aerosol layer illuminated by a monodirectional beam [*Coakley and Chylek*, 1975; *King and Harshvardhan*, 1986]

$$R(\mu_0) = \frac{\tau\omega\beta(\mu_0)}{\mu_0} \qquad (3)$$

$$T(\mu_0) = 1 - \frac{\tau}{\mu_0}[1-\omega+\omega\beta(\mu_0)] \qquad (4)$$

where μ_0 is a direction cosine of incoming radiation with respect to the normal to the layer, τ is the extinction optical depth ($\tau = \tau_{sc} + \tau_{abs}$, where τ_{sc} and τ_{abs} are optical depths due to scattering and absorption, respectively), $\omega = \tau_{sc}/\tau$ is a single scattering albedo and $\omega\beta(\mu_0)$ is a fraction of radiation scattered into the upper hemisphere.

The total reflectance (spherical albedo) R and total transmittance T of a layer is obtained by integration of equations (3) and (4) over all angles of incoming radiation [*Coakley*

and Chylek, 1975; *King and Harshvardhan*, 1986]. The absorbance $A = 1 - R - T$. We obtain

$$R = 2\tau\omega\beta = 2\beta\tau_{sc} \quad (5)$$

$$A = 2\tau(1-\omega) = 2\tau_{abs} \quad (6)$$

where $\beta = \int_0^1 \beta(\mu_0)d\mu_0$.

When the aerosol layer is placed over a surface with the surface albedo a, the albedo R' of the combined system is affected by multiple reflections between the surface and the aerosol layer. The two stream approximation for incident isotropically diffused radiation [*Chylek and Coakley*, 1974] can be used to obtain the albedo R' of a combined system. The change in albedo ΔR is defined as the difference between the albedo R', of the combined surface-aerosol system, and the original albedo a. Using the equation (6) of Chylek and Coakley (1974) with an appropriate expansions for an optically thin layer we obtain

$$\Delta R = (1-a)^2 R - 2aA = (1-a)^2 2\beta\tau_{sc} - 4a\tau_{abs} \quad (7)$$

Thus the eq. (1) is modified to a more general form valid also for absorbing aerosols

$$\Delta F_R = -\frac{S_0}{4} T_{atm}^2 (1-N)[(1-a)^2 2\beta\tau_{sc} - 4a\tau_{abs}] \quad (8)$$

For a nonabsorbing aerosol we have $\tau_{abs} = 0$ and the equation (8) reduces to the eq. (1) used by Charlson et al. (1992) for the case of nonabsorbing sulfate aerosols. However, for absorbing aerosols the second term cannot be, generally, neglected.

Although the aerosol direct forcing can be determined by solving a more accurate form of radiation transfer model [*Coakley et al.*, 1983], the advantage of an analytical solution in the form of eq. (8) is an explicit dependence on the individual parameters determining the forcing. We note that the first term (the scattering term) on the right hand side of eq. (7) depends on the aerosol upper hemispheric scattering fraction, β, while the the second term is independent of β.

Climate Forcing of Smoke Aerosols

The upscattered fraction β is a sensitive function of particle size. It is equal to 0.5 for small particles in the region of Rayleigh scattering and it decreases with increasing size of aerosol particles. The choice of particle size distribution and the corresponding value of the upper hemispheric scattering fraction is crucial for the estimate of aerosol direct climate forcing.

Aerosol particles produced by biomass burning have a wide range of sizes. Aerosol particles produced by fires will grow at ambient relative humidity to larger equilibrium sizes. The range of observed particle radii [*Mulholland and Ohleimiller*, 1982; *Helsper et al.*, 1980; *Woods et al.*, 1991; *Cachier et al.*, 1991; *Einfield et al.*, 1991] is from 0.01μm to about 5μm. Radke et al. (1991) observe three distinct modes of biomass burning aerosols: nucleation mode with radii below 0.05μm, accumulation mode with radii $0.05\mu\text{m} < r < 1.0\mu\text{m}$ and coarse mode with radii above 1μm. The accumulation mode dominates aerosol mass and aerosol light scattering properties [*Radke et al.*, 1991]. Similar distribution of sizes is found by Holben et al. (1991) who use the log-normal size distribution to describe the biomass burning aerosols.

To investigate the dependence of aerosol direct forcing on the aerosol size distribution, we use log-normal size distribution of the form

$$n(r) = \frac{N}{\sqrt{2\pi}\, r \ln\sigma} \exp[-\frac{(\ln r - \ln r_0)^2}{2\ln^2\sigma}] \quad (9)$$

with the geometric mean radius, r_0, between 0.05 and 0.30μm and with standard deviation $\ln\sigma = 0.7$. The effective radius ($r_{eff} = r_0 \exp[2.5\ln^2\sigma]$) of these distributions changes from 0.17μm to 1.02μm, which is within the range of the observed effective radii from fresh to aged biomass burning aerosols as given by Holben et al. (1991) and Radke et al. (1991).

We used several refractive indices with real part between 1.33 and 1.43 to calculate the asymmetry parameter, g, averaged over the considered log-normal size distributions. The dependence on the refractive index (within the stated limits) was found weak compared to the dependence on the effective radius of the size distribution.

The upscattering fraction is calculated using an approximate relation $\beta = (1 - g/2)/2$ [*Sagan and Pollack*, 1967] which for considered sizes of aerosol particles is close to the upscattering fraction for an average 60° solar zenith angle [*Wiscombe and Grams*, 1976]. For the smallest fresh biomass burning aerosols ($r_0 = 0.05\mu$m) we obtain $\beta = 0.37$, while for the large size end of the aged biomass burning aerosols ($r_0 = 0.30\mu$m) we have $\beta = 0.11$.

To isolate the effect of smoke aerosol size distribution on the climate forcing we take all other parameters as in Penner et al. (1992). Thus we have the absorption and the scattering optical thickness of a smoke layer $\tau_{abs} = 0.0026$ and $\tau_{sc} = 0.030$, the global averaged albedo $a = 0.22$ over the land and $a = 0.06$ over the ocean with 80% of aerosols being over the land; the solar constant of $1370 W/m^2$, the atmospheric transmittance is taken to be $T_{atm} = 0.79$ [*Penner et al.*, 1992] and cloudiness $N = 0.6$.

The results are summarized in Table 1. The absorption term in eq. (8) reduces the estimated cooling effect by about $0.17 W/m^2$ for the considered absorption optical thickness

Table 1. Effective radius r_{eff} and the upscattered fraction β of the log normal size distribution with $\ln\sigma = 0.7$ and the given mean geometric radius r_0

$r_0(\mu\text{m})$	$r_{eff}(\mu\text{m})$	β	ΔF
0.05	0.17	0.37	-1.1
0.06	0.20	0.35	-1.0
0.08	0.27	0.31	-0.9
0.10	0.30	0.28	-0.8
0.15	0.51	0.22	-0.6
0.20	0.68	0.18	-0.4
0.25	0.85	0.14	-0.3
0.30	1.02	0.11	-0.2

The direct radiative forcing $\Delta F(W/m^2)$ (given by eq. (8)) induced by an aerosol layer of optical thickness $\tau_{sc} = 0.03$, $\tau_{abs} = 0.0026$ composed of aerosol particles described by the considered size distribution varies between -0.2 and $-1.1 W/m^2$

of 0.0026. This absorption term does not depend explicitly on the aerosol size distribution. We obtain the biomass burning induced direct climate forcing ΔF ranging from about $-0.2W/m^2$ to $-1.1W/m^2$ depending on the size distribution of considered aerosols.

We have repeated our calculations for log-normal size distributions with $\ln\sigma = 0.5$ and 0.3. The most important parameter determining the upscattering fraction, β, is an effective radius of the size distribution. Size distributions with different values of r_0 and $\ln\sigma$, however, with the same value of effective radius, r_{eff}, will give similar values of β. This is consistent with the effective radius being the most suitable variable characterizing the scattering properties of particle polydispersion [*Hansen and Travis*, 1974; *Damiano and Chylek*, 1994].

Conclusion and Discussion

We have shown that the direct radiative forcing of an optically thin aerosol layer is given by eq. (8). The advantage of expression (8) is its simplicity and an explicit dependence of the forcing on aerosol layer optical thickness and aerosol upscattering fraction. For the case of nonabsorbing aerosols the eq. (8) is reduced to the form used previously by Charlson et al. (1992) and Penner et al. (1992).

We have shown that the aerosol upscattering fraction is a sensitive function of aerosol size distribution. Considering the measurements of aerosol size distributions produced by biomass burning [*Mulholland and Ohleimiller*, 1982; *Helsper et al.* , 1980; *Woods et al.* , 1991; *Cachier et al.* , 1991; *Einfield et al.* , 1991; *Radke et al.* , 1991; *Holben et al.* , 1991], the direct radiative forcing due to smoke aerosols varies between $-0.2W/m^2$ and $-1.1W/m^2$ depending on the size distribution used. A similar dependence on size distribution of radiative effects of sulfate aerosols was pointed out by Kiehl and Briegleb (1993). This range of uncertainty can be reduced by additional field measurements and modeling of the smoke aerosol size distribution and its changes with relative humidity. The result of Penner et al. (1992) of radiative forcing $-0.8W/m^2$ (when absorption was accounted for) is reproduced with the size distribution parameters $r_0 = 0.09\mu$m and $\ln\sigma = 0.7$.

We have limited our consideration to the clear sky portion of the atmosphere in a similar way as has been done by Charlson et al. (1992) and Penner et al. (1992). Coakley et al. (1983) considered climate forcing of tropospheric aerosols including the cloudy part of the atmosphere. His results suggest that inclusion of aerosol forcing over the cloudy part of the atmosphere increases the direct aerosol forcing by 20 to 25%.

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