Soot and smoke aerosol may not warm climate

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[1] Soot and smoke aerosol contain black carbon, which absorbs solar radiation. These aerosols may reduce the overall negative climate forcing of anthropogenic aerosols by absorbing radiation that might otherwise be scattered back to space. They may also reduce overall cloudiness, an effect termed the "semidirect" effect, which is thought to enhance climate warming. Here, we evaluate the climate forcing associated with black carbon and other aerosols using the concept of "relaxed forcing," which is the forcing associated with two simulations using fixed sea surface temperatures. The consideration of longwave perturbations associated with the relaxed forcing leads to a diminished or even negative semidirect effect associated with absorbing aerosols rather than an enhanced warming. The overall forcing depends significantly on the altitude of injection of the aerosols because higher-altitude injections tend to enhance the negative longwave forcing. In addition, high-altitude injection of absorbing aerosols can increase cloudiness at lower altitudes where temperatures, in general, may decrease. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 1610 Global Change: Atmosphere (0315, 0325); 3309 Meteorology and Atmospheric Dynamics: Climatology (1620); *KEYWORDS:* soot, biomass, aerosol

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1. Introduction

[2] Black carbon (the optically absorbing component of soot and smoke) is produced during the incomplete combustion of various fuels, with the most important sources being fossil fuel combustion and biomass burning [*Turco et al.*, 1983; *Penner et al.*, 1992, 1993; *Liousse et al.*, 1996; *Cooke et al.*, 1999]. Black carbon (BC) is a major light absorbing aerosol species within the atmosphere, and its presence within aerosol is known to be important to climate. Moreover, because of its short lifetime relative to that of CO_2 it has been suggested that controls of black carbon may provide a short-term solution for global warming that is easier to achieve than controls on CO_2 [*Hansen et al.*, 2000; *Jacobson*, 2002].

[3] The direct forcing by fossil fuel black carbon and organic matter (OM) associated with fossil fuel BC emissions has been estimated to range from +0.16 to +0.42 Wm^{-2} [*Cooke et al.*, 1999; *Haywood et al.*, 1997; *Myhre et al.*, 1998; *Penner et al.*, 1998; *Haywood and Ramaswamy*, 1998; *Jacobson*, 2002] and the total absorbed radiation associated with these components has been estimated to range from 0.56 to about 2 Wm^{-2} [*Penner et al.*, 1998; *Ramanathan et al.*, 2001]. Here, we adopt the term "fossil fuel soot" as a shorthand for fossil fuel BC+OM. Biomass burning aerosols, which contain about 10% BC by mass, have been estimated to lead to a direct forcing of −0.16 to −0.74 Wm^{-2} [*Penner et al.*, 1998; *Hobbs et al.*, 1997] while the absorbed radiation was estimated to range from 0.75 to about 2 Wm^{-2} [*Penner et al.*, 1998; *Ramanathan et al.*, 2001].

[4] The scattering part of biomass smoke and fossil fuel soot is primarily composed of organic matter. In addition other, mostly scattering, trace constituents make up as much as 15% of the total mass of biomass smoke [*Liousse et al.*, 1996]. These components, together with the water soluble organic and inorganic constituents that are emitted as part of the smoke and soot, the soluble components attached through coagulation, and the soluble components formed by oxidation allow the particles to act as effective cloud condensation nuclei (CCN) [*Liousse et al.*, 1996; *Jacobson*, 2001; *Decesari et al.*, 2002]. Thus emissions of BC (and OM) also participate in the indirect effect of aerosol particles on climate wherein aerosol particles may increase droplet concentrations and the albedo of clouds. While sulfate aerosols also participate in the indirect effect, because most of the sulfate in aerosols is formed in clouds through aqueous chemical reactions that do not form new particles, the indirect effect of biomass smoke and fossil fuel soot may be responsible for more than 80% of the total (e.g., biomass smoke, fossil fuel soot, and anthropogenic sulfate) indirect effects of anthropogenic aerosols on climate [*Chuang et al.*, 2002].

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Table 1. Annual Average Aerosol Burden for the Different Aerosol Types Included in the Simulations

Aerosol Type	NH, Tg	SH, Tg	Global, Tg
Anthropogenic SO_4^{2-}	0.87	0.22	1.09
Natural SO_4^{2-}	0.45	0.42	0.86
Fossil Fuel OC	0.39	0.03	0.41
Fossil Fuel BC	0.08	0.01	0.09
Biomass OC	1.28	1.24	2.52
Biomass BC	0.13	0.13	0.26
Biomass OC, surface[a]	0.49	0.52	1.02
Biomass BC, surface[a]	0.05	0.06	0.11
Natural OC	0.13	0.10	0.23
Dust ($r < 1$ μm)	11.11	3.57	14.68
Sea salt ($r < 1$ μm)	1.82	2.85	4.68

[a]Biomass aerosols were injected at the surface in this simulation.

[5] In addition to these direct and indirect effects of smoke and soot, BC may have another effect: by locally warming the atmosphere, the presence of BC can reduce cloud cover and liquid water path, leading to a decrease in the reflected radiation. In previous studies, this so-called "semidirect" effect was thought to add to the warming caused directly by black carbon absorption thereby enhancing its warming impact on climate [*Hansen et al.*, 2000; *Ackerman et al.*, 2000; *Lohmann and Feichter*, 2001].

[6] Here, we evaluate the effects of soot and smoke using the GRANTOUR/CCM global climate model [*Taylor and Penner*, 1994]. This model has been enhanced by including a calculation of the effect of BC in cloud droplets on the albedo of clouds [*Chuang et al.*, 2002]. This feature allows an accurate calculation of the heating by BC together with the indirect effect of smoke and soot aerosols. In the presence of soot, increasing droplet concentrations associated with the indirect effect may decrease the reflection of solar radiation if the clouds are sufficiently thick [*Twomey*, 1977].

2. Methods

[7] Our calculation of the radiative effects of smoke and soot is based on a prognostic calculation of the aerosol mass concentration of biomass aerosols, fossil fuel black carbon and organic carbon aerosols, sulfate, sea salt, dust, and natural organic aerosols. We use the specification of emissions from the IPCC workshop for natural and carbonaceous aerosols, [*Penner et al.*, 2001] with separate treatments for the emissions of biomass smoke and fossil fuel soot. Fossil fuel emissions are injected into the model in the lowest 100 hPa, but the emissions from biomass burning are injected between 400 and 700 hPa to reproduce the altitude of the main smoke layers observed during the SAFARI campaign [*Anderson et al.*, 1996]. The calculated global aerosol burdens and vertical profiles are given in Table 1 and Figure 1, respectively. We also examine simulations in which biomass aerosols are injected in the lowest 100 hPa. In these simulations, their removal by both precipitation and dry deposition is much more efficient, and the total burden decreases significantly. The paper by *Chuang et al.* [2002] evaluated the comparison of our model-predicted BC and organic carbon (OC) with surface observations for the case of surface injection of both smoke and soot aerosols. Here, our midtroposphere injection of biomass smoke provides a better estimate of the vertical profiles measured during the SAFARI campaign, but does not significantly degrade our comparison with surface observations (S. Zhang, University of Michigan, Ph.D. thesis in preparation, 2003).

[8] The calculation of the direct radiative forcing of biomass smoke uses the radiative treatment described by *Grant et al.* [1999] for aerosol size distributions observed in plumes off of Africa [*Anderson et al.*, 1996] while fossil fuel OM and BC is treated as an internal mixture with the size distribution specified by *Penner et al.* [1998]. The calculation of the change in cloud droplet number or the "first indirect effect" approximately accounts for the separate effects of sulfate aerosol formed in cloud and that formed by the homogeneous reaction of SO_2 with OH in air as well as the effects of adding smoke and soot aerosol to preindustrial aerosols [*Chuang and Penner*, 1995]. Some general circulation model (GCM) calculations have also included the effects of changes in cloud droplet number on the precipitation efficiency (and thus the lifetime) of clouds. The forcing from this so-called "second indirect effect" has varied between about 35% of that from the first indirect effect to approximately equal to it in different models [*Penner and Rotstayn*, 2000]. The increased negative climate forcing results from increases in cloud liquid water path and cloud fraction. However, empirical evidence on a global scale does not support a change in low-altitude liquid water path when aerosols increase [*Nakajima et al.*, 2001] (though some small-scale observations support this increase [*Rosenfeld*, 2000]), and the global forcing associated with this impact was not estimated by IPCC because of its large uncertainty [*Ramaswamy et al.*, 2001]; hence it is not included here.

[9] We calculate the individual and combined indirect plus direct forcing associated with fossil fuel soot and biomass smoke as well as that associated with the sum of anthropogenic sulfate, fossil fuel soot, and biomass smoke.

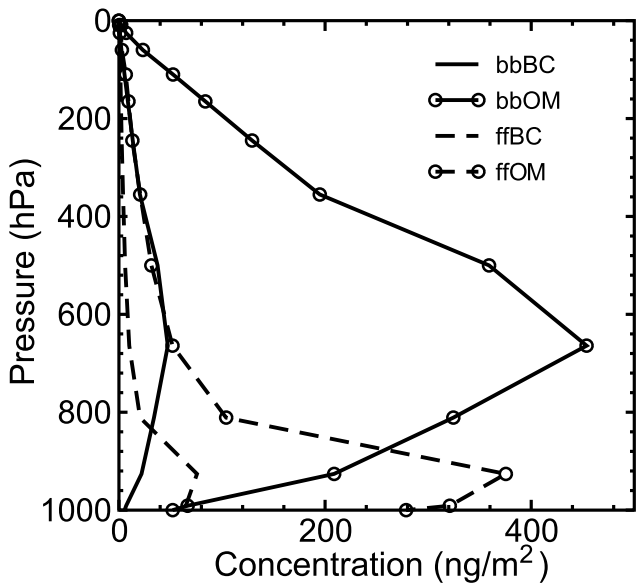

Figure 1. Global and annual average change in BC and organic matter (OM) concentration associated with the fossil fuel soot and biomass smoke perturbations.

Because organic carbon aerosols are coemitted with black carbon during the same combustion processes [*Cooke et al.*, 1999] we believe it is inappropriate to consider either BC or organic matter independently. Thus our analysis differs somewhat from those of recent articles that consider black carbon forcing independent of its association with organic matter [*Hansen et al.*, 2000; *Jacobson*, 2001]. In all simulations, the preindustrial or baseline case is that determined using the dust, sea salt, and natural organic and sulfate aerosol distributions from emissions specified in the IPCC model intercomparison workshop [*Penner et al.*, 2001].

[10] We examine the semidirect effects of smoke and soot using the concept of relaxed forcing (termed "quasi-forcing" by *Rotstayn and Penner* [2001]). Relaxed forcing has commonly been used to evaluate the change in top of the atmosphere radiative fluxes associated with changes in the precipitation efficiency of clouds caused by increases in aerosols (the second indirect effect). In this calculation, forcing is estimated as the difference between two multi-year simulations with fixed sea surface temperature so that aerosols are allowed to change the air and land temperatures, water vapor, and cloud fields through their direct radiative effects and through their indirect cloud microphysical effects. This differs from a pure or "instantaneous" calculation of forcing which uses two radiation calls at each time step of the GCM calculation to compute the forcing, thereby guaranteeing that the only changes in the calculation of forcing are those specified by the perturbed quantity. Relaxed forcing is therefore not a pure forcing since it includes the "feedbacks" of the climate that adjust on the fast timescales associated with changes in land surface temperatures, lapse rate, water vapor profiles, and clouds when SST's are held fixed but aerosols and their effects on clouds are allowed to change. We note that the "adjusted forcing" used by many researchers is also not a pure forcing. In the adjusted forcing, the change in radiative fluxes associated with the change in stratospheric temperatures (that results from an imposed change in gas or aerosol concentrations) as well as the change in radiative fluxes associated directly with the imposed change in gas or aerosol concentrations is included in the calculation of forcing (see *Hansen et al.* [1997] for a discussion of instantaneous or pure forcing and adjusted forcing). The calculation of the adjusted forcing requires two radiation calls, one with the perturbed stratospheric temperature saved from a simulation where the temperature is allowed to respond to the changes in gas or aerosol concentrations and one with the unperturbed stratospheric temperatures and gas or aerosol concentrations. Thus the adjusted forcing is also a type of "relaxed" forcing, but it only allows the relaxation of the stratospheric temperatures (which adjust on a 1 month to 1 year timescale) in the calculation of forcing. In contrast, the relaxed forcing defined here also includes changes in tropospheric air temperature, land surface temperature, water vapor and cloud fields.

[11] The adjusted forcing has been adopted as a better measure of the equilibrium global average surface temperature change because it maintains an approximately proportional relationship between the forcing and the global average surface temperature change at equilibrium [*Ramaswamy et al.*, 2001]. *Rotstayn and Penner* [2001] showed that the relaxed forcing is also a better measure of the equilibrium global average surface temperature change when the cloudiness changes associated with the second indirect effects of sulfate aerosols are included. Moreover, they showed that a calculation of the instantaneous forcing (which is close to the adjusted forcing for aerosols with effects that do not cause absorption of radiation in the stratosphere) and a calculation of the relaxed forcing were within 20% for the first indirect effect of sulfate aerosols. The calculation of the instantaneous forcing is possible for this case because only the effective radius of the cloud changes while cloud amounts are constant. Thus two radiation calls with different effective radii for the clouds can be performed. The relaxed forcing for $2 \times CO_2$ (which does change stratospheric temperatures) was within 6% of the adjusted forcing for $2 \times CO_2$ (3.31 Wm^{-2} versus 3.51 Wm^{-2}, respectively). In simulations where cloudiness changes as a result of the second indirect effect, it is not possible to determine a pure or instantaneous forcing since this forcing must by its nature include changes to cloud amounts. For this case, they examined the climate sensitivity parameter, which quantifies the ratio of the global average surface temperature change and the global average forcing and thus measures its proportionality. This only varied from 0.69 to 0.87°K/(W m^{-2}) for the range of aerosol and greenhouse gas simulations that used relaxed forcing rather than the instantaneous forcing for sulfate aerosols or the adjusted forcing for $2 \times CO_2$ [*Rotstayn and Penner*, 2001]. The climate sensitivity parameter would have been as large as 1.66°K/(W m^{-2}) if the instantaneous forcing rather than the relaxed forcing estimates had been used to estimate the climate sensitivity for the second indirect effect of sulfate aerosols. Thus these studies suggest that the "relaxed forcing" determined from the difference between 2 multiyear simulations with fixed sea surface temperature can be used to approximately project the steady state average surface temperature change from a simple proportionality between temperature and forcing when changes in cloudiness are part of the perturbation.

[12] *Hansen et al.* [1997] showed that a proportional response between the instantaneous or adjusted forcing and the global average surface temperature change is also not possible for absorbing aerosols because of changes to clouds and changes to the vertical temperature structure. Because the use of relaxed forcing maintained an approximately proportional response between the forcing and the global average surface temperature change in the study of *Rotstayn and Penner* [2001], it may allow the continued use of the concept of "forcing" as an approximate climate predictor for different forcing mechanisms. Here, we use the relaxed forcing to examine the effect of cloudiness changes caused by BC absorption on radiative fluxes. Future work will consider the effects of BC on the predicted equilibrium temperature changes.

3. Results

[13] We examined the radiative changes associated with the direct, semidirect, and indirect effects of aerosols using the relaxed forcing methodology outlined above to evalu-

Table 2. Global and Annual Average Change in Net Incoming Radiation and Change in Liquid Water Path[a]

Case	Instantaneous Shortwave Forcing,[b] W/m^2	Adjusted Forcing, W/m^2	Change in Convective Water Path,[c] g/m^2	Change in Stratiform Water Path,[c] g/m^2	Shortwave Relaxed Forcing, W/m^2	Longwave Relaxed Forcing, W/m^2	Total Relaxed Forcing, W/m^2	Total Semidirect Effect, W/m^2
Fossil fuel, direct	0.17	0.14	−0.20 (0.20)	−0.09[d] (0.26)	0.28 (0.32)	−0.21 (0.17)	0.07[d] (0.39)	−0.09[d] (0.39)
Biomass, direct	−0.03	−0.10	−0.75 (0.30)	−0.22 (0.27)	0.01[d] (0.29)	−0.39 (0.37)	−0.38 (0.46)	−0.36 (0.47)
Biomass, surface,[e] direct[f]	−0.13	−0.16	−0.34 (0.24)	−0.07[d] (0.17)	0.00[d] (0.22)	−0.15 (0.14)	−0.15 (0.20)	−0.03[d] (0.20)
Fossil + biomass, direct	0.14	0.04	−0.98 (0.16)	−0.28 (0.16)	0.34 (0.22)	−0.57 (0.17)	−0.23 (0.37)	−0.37 (0.37)
All (fossil + biomass + sulfur), direct	−0.90	−1.01	−0.88 (0.19)	−0.29 (0.28)	−0.82 (0.36)	−0.41 (0.43)	−1.24 (0.35)	−0.34 (0.36)
All, surface,[e] direct	−1.01	−1.04	−0.55 (0.18)	−0.20 (0.22)	−0.80 (0.36)	−0.11[d] (0.28)	−0.91 (0.23)	0.10[d] (0.23)
Fossil fuel, indirect + direct	−0.05	−0.07	−0.25 (0.22)	−0.13 (0.20)	0.12[d] (0.30)	−0.11[d] (0.25)	0.01[d] (0.25)	0.06[d] (0.25)
Biomass, indirect + direct	−1.21	−1.28	−0.67 (0.17)	−0.13[d] (0.29)	−1.41 (0.52)	−0.31 (0.18)	−1.71 (0.44)	−0.50 (0.44)
Biomass, surface,[e] indirect + direct[f]	−0.76	−0.77	−0.29 (0.20)	0.00[d] (0.31)	−0.78 (0.55)	−0.04[d] (0.29)	−0.82 (0.42)	−0.06[d] (0.41)
Fossil + biomass, indirect + direct	−1.21	−1.30	−0.89 (0.20)	−0.30 (0.26)	−1.18 (0.36)	−0.43 (0.26)	−1.61 (0.37)	−0.39 (0.38)
All, indirect + direct	−2.40	−2.52	−0.82 (0.15)	−0.18 (0.22)	−2.71 (0.42)	−0.19 (0.16)	−2.90 (0.41)	−0.50 (0.41)
All, surface,[e] indirect + direct	−1.97	−2.01	−0.50 (0.21)	−0.14 (0.23)	−1.92 (0.32)	−0.06[d] (0.20)	−1.98 (0.33)	−0.01[d] (0.34)

[a]The changes are the average from a 10-year simulation of the GCM. Values in parentheses are the standard deviation.
[b]The pure (instantaneous) longwave forcing is less than 0.01 Wm^{-2}.
[c]These are estimated changes in the spatial and time-averaged liquid water path. These changes include both changes in in-cloud liquid water content and cloud fraction.
[d]Numbers are not significantly different from zero at the 5% confidence level.
[e]Biomass aerosols were injected at the surface in this simulation.
[f]These simulations were 9 years in duration.

ate the total forcing associated with smoke and soot aerosols. Cloudiness changes were evaluated as the time-averaged difference in in-cloud liquid water path times the cloud fraction between the perturbed and preindustrial cases.

[14] We considered the direct and the indirect plus direct forcing for 10 year long simulations of four different combinations of perturbations: fossil fuel soot, biomass smoke, the combined forcing by fossil fuel soot and biomass smoke, and the combined forcing by anthropogenic sulfate, fossil fuel soot and biomass smoke aerosols. In addition we considered two sensitivity cases for biomass smoke (i.e., for the biomass smoke only case and the combined sulfate, fossil fuel soot, and biomass smoke case) in which we emit them near the surface. Table 2 shows the top of atmosphere (TOA) instantaneous forcing, the global annual mean change in liquid water path for stratiform and convective clouds, and the relaxed forcing broken into its shortwave and longwave components. For biomass aerosols injected above the surface we find important changes to the stratospheric temperatures. Hence we include an estimate of the adjusted forcing in the second column of Table 2. This was calculated using a single column model to estimate the change in longwave forcing associated with the change in the global average stratospheric temperature profile. The sum of this longwave forcing and the instantaneous (shortwave) forcing provides an estimate of the adjusted forcing.

[15] The semidirect effect is approximated by the difference between the total relaxed forcing and the instantaneous calculation of forcing. Thus this method for quantifying the semidirect effect isolates the forcing changes associated with the changes in air and land surface temperatures and the changes in cloudiness caused by these temperature changes from the changes due to the direct radiative effects of the aerosols themselves and the indirect radiative effects caused by changing cloud droplet radii. For the semidirect effect, the change in the TOA longwave radiation reflects both the change in the vertical temperature profile as well as the change in cloudiness and precipitable water. Shortwave TOA changes mainly reflect the change in BC and OM aerosol concentrations as well as the changes in the time-averaged stratiform cloud liquid water path (Figure 2).

[16] Table 2 lists the estimated instantaneous, adjusted, relaxed, and semidirect forcings for our calculations. The calculation for fossil fuel soot direct shortwave TOA instantaneous forcing is 0.17 Wm^{-2}, but the shortwave relaxed forcing is larger, 0.28 ± 0.32 Wm^{-2} (see Table 2). (Instantaneous forcings have a standard deviation in our calculations of less than 0.02 Wm^{-2}; hence all numbers are reported to 2 significant digits here.) While our reported relaxed forcings are often slightly less than or about equal to the standard deviation of the annual average changes calculated over the 10 years of simulation, most are significantly different from zero within a 5% confidence interval according to the standard one-tailed *t* test (Table 2 indicates those forcings that are not significantly different from zero by listing them in italics). Moreover, the reported changes in longwave and shortwave forcing are consistent with the 10-year average change in the global average temperature profile, the liquid water path, and the water vapor profile for

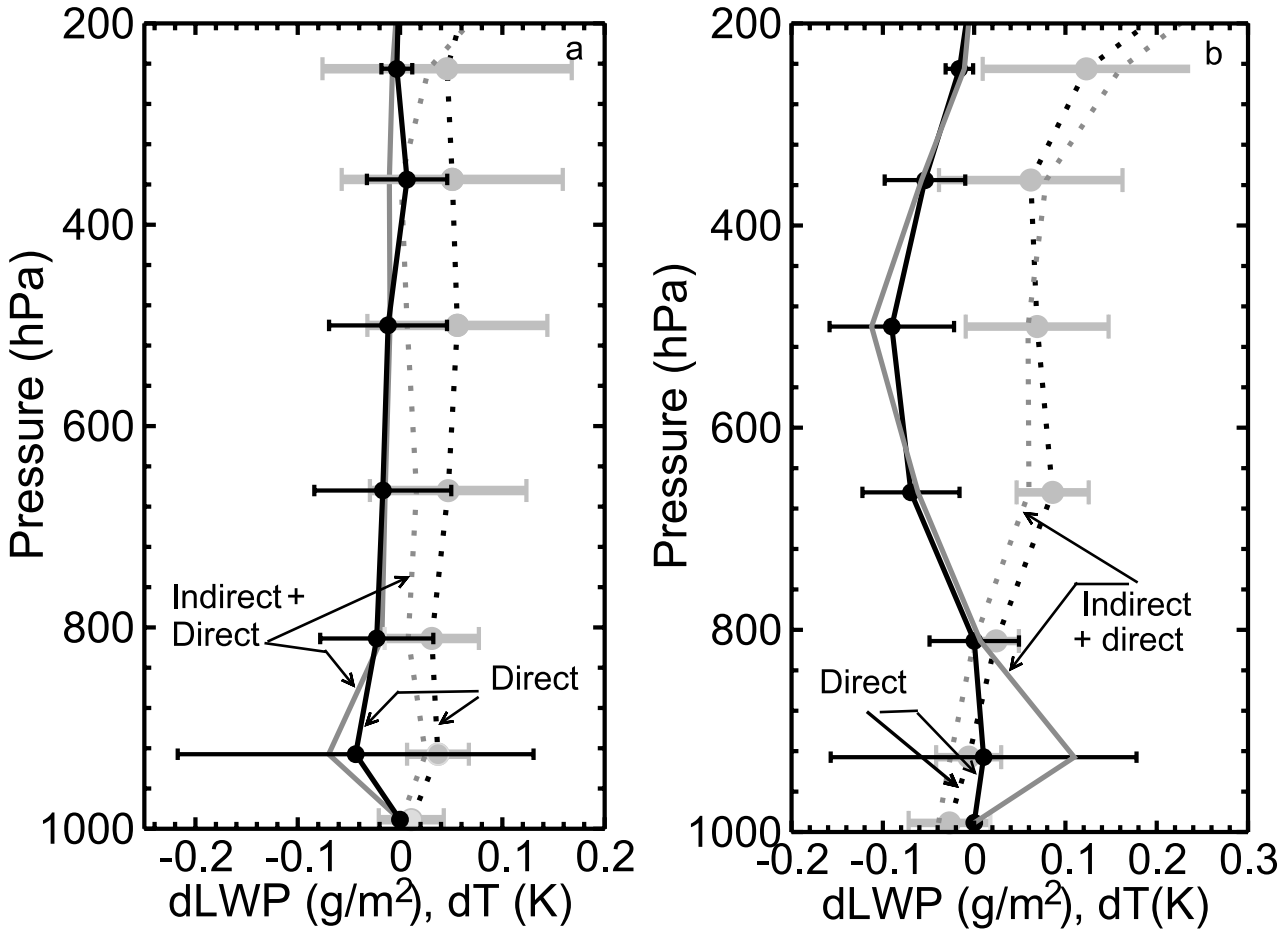

Figure 2. Global and annual average stratiform liquid water path change in each level of the climate model (solid curves) and global annual average temperature change (dotted curves) associated with (a) fossil fuel soot (BC + OM) direct and indirect plus direct effects and (b) biomass smoke direct and indirect plus direct effects. The black horizontal bars show the standard deviation associated with the liquid water path changes for the direct effect, and the grey horizontal bars show the standard deviation associated with the temperature changes for this case.

both the longwave changes (see Table 3) and the shortwave changes (not shown). For example, the calculated difference between the shortwave relaxed forcing and the shortwave TOA instantaneous forcing for direct forcing by fossil fuel soot is consistent with the change in stratiform liquid water path of -0.09 gm^{-2} despite the fact that this change in liquid water path is not significantly different from zero. We conclude that the relaxed forcings that we calculate are physically sensible. Moreover, because the time series of changes are stationary for the entire 10 years of simulation, we expect that our central estimates in Table 2 are unlikely to change with longer simulations.

Table 3. TOA Longwave Relaxed Forcing Estimated Using A Single-Column Radiation Model and the Global Average Changes for the Simulations Shown in Table 2[a]

Case	dLW(Tg), Wm^{-2}	dLW(T3d), Wm^{-2}	dLW(H_2O), Wm^{-2}	dLW(LWP), Wm^{-2}	dLW(All), Wm^{-2}
Fossil fuel, direct	0.00	−0.17	0.04	−0.04	−0.17
Biomass, direct	0.03	−0.27	0.04	−0.14	−0.34
Fossil + biomass, direct	0.04	−0.45	0.13	−0.20	−0.48
All (fossil + biomass + sulfur), direct	0.08	−0.34	0.04	−0.18	−0.39
All, surface,[b] direct	0.07	−0.06	−0.03	−0.09	−0.11
Fossil fuel, indirect + direct	0.01	−0.10	0.02	−0.04	−0.11
Biomass, indirect + direct	0.04	−0.26	0.06	−0.15	−0.31
Fossil + biomass, indirect + direct	0.06	−0.30	0.03	−0.20	−0.40
All, indirect + direct	0.13	−0.23	0.01	−0.16	−0.26
All, surface,[b] indirect + direct	0.08	−0.04	−0.06	−0.13	−0.14

[a]Shown are the changes in radiation due to changes in surface temperature (Tg), atmospheric temperature profile (T3d), precipitable water profile (H_2O), liquid water path profile (LWP), and all changes acting together (all).
[b]Biomass aerosols were injected at the surface in this simulation.

Therefore, because the t values for the standard student's *t* test are expected to decrease over time, we conclude that all of the estimated changes in Table 2 may eventually become statistically significant.

[17] We note that although the change in liquid water path associated with convective clouds for fossil fuel soot is larger than that associated with stratiform clouds, the former changes have little impact on the change in net incoming shortwave flux because they are optically much thicker than the stratiform clouds. We verified this statement by running an off-line shortwave radiative code using the global average in-cloud convective cloud liquid water path change and the in-cloud stratiform liquid water path change to estimate the shortwave forcing associated with these changes in liquid water path. The shortwave forcing associated with changes in convective clouds was only 0.01 Wm^{-2}, while that associated with stratiform liquid water changes was 0.12 Wm^{-2}. At the same time, however, there is a change in the longwave relaxed forcing of -0.21 ± 0.17 Wm^{-2}, which is mainly associated with the slight positive temperature change (Figure 2a and Table 3). The change in the longwave forcing due to stratospheric temperature changes alone in this case is estimated as -0.03 Wm^{-2} from the single column radiative transfer model (the difference between column 2 and column 1 in Table 2). Thus the total shortwave plus longwave relaxed forcing, 0.08 ± 0.39 Wm^{-2}, is smaller than the instantaneous calculation of forcing, 0.17 Wm^{-2}. When we evaluate the semidirect effect as the difference between the relaxed forcing and the instantaneous calculation of forcing, it is small and negative, -0.09 ± 0.39 Wm^{-2}, because of the negative longwave relaxed forcing. Thus the relaxed forcing estimate suggests a diminished warming by fossil fuel soot compared to the instantaneous (or adjusted) forcing.

[18] For biomass smoke, the TOA shortwave forcing is nearly the same whether calculated as an instantaneous forcing or as a relaxed forcing (-0.03 Wm^{-2} for the instantaneous forcing calculation compared to 0.01 ± 0.29 Wm^{-2} when calculated as a relaxed forcing). The increase in shortwave forcing associated with the relaxed forcing by biomass smoke is much smaller than that associated with fossil fuel soot even though the change in stratiform liquid water path, -0.22 ± 0.27 gm^{-2}, is more than 2 times larger than the change associated with fossil fuel soot aerosols, i.e., -0.09 ± 0.26 gm^{-2}. This difference is associated with the injection of biomass smoke at midtroposphere levels. The higher-altitude injection of biomass smoke causes an increase of temperature near the layer of the main injection height, but a decrease in temperature below about 800 hPa. The decrease in temperature at the surface and the decrease in atmospheric lapse rate, in turn, lead to a slight increase in the stratiform liquid water path below 800 hPa (Figure 2b). The decreased cloudiness at higher altitudes for the biomass case has a smaller effect on the outgoing shortwave flux than does the decrease in low-altitude cloud for the fossil fuel soot case (which was verified, using our single column radiative model). This is because when higher-altitude clouds decrease, the solar flux can still be reflected back to space by lower-altitude clouds. If, however, there is mainly a decrease in clouds at lower altitudes as in the fossil fuel case, a large fraction of the solar flux penetrates this layer and is absorbed by the surface (the average surface albedo is 0.15–0.20).

[19] The change in longwave radiation associated with the direct effects of biomass smoke is about twice as large as the longwave relaxed forcing of fossil fuel soot (i.e., -0.39 ± 0.37 Wm^{-2} instead of -0.21 ± 0.17 Wm^{-2}), mainly because the change in temperature for the biomass smoke perturbation is much larger near the effective emitting level of the atmosphere, for example, 700 hPa (see Figure 2). The change in the longwave forcing due to stratospheric temperature changes alone in this case is estimated as -0.07 Wm^{-2} from the single column radiative transfer model. The relatively large change in longwave radiation for biomass smoke is also associated with the fact that the cloudiness changes occur mainly near 500 hPa (Figure 2b), and these changes in high-altitude clouds have a larger impact on longwave radiation than do changes in clouds near the surface. As a result, the total (shortwave and longwave) relaxed forcing is -0.38 ± 0.46 Wm^{-2}, and the semidirect effect caused by the decrease in cloudiness is relatively large and negative, -0.36 ± 0.47 Wm^{-2}.

[20] Table 3 shows the separate contributions to the total change in the longwave radiation from changes to the global average surface temperature, the atmospheric temperature profile, the precipitable water profile, and the liquid water path profile. For the biomass smoke and fossil fuel soot calculations, the negative longwave relaxed forcing is mainly associated with the temperature perturbation. The change in the longwave forcing due to the change in liquid water path is between 20% and 64% as large as the longwave relaxed forcing associated with the change in atmospheric temperature. A smaller, positive change in longwave relaxed forcing is associated with the change in precipitable water. These calculations also show that the negative longwave relaxed forcing associated with cloudiness changes contributes significantly to the total relaxed forcing for biomass smoke injected into the middle troposphere. *Hansen et al.* [1997] studied the addition of absorbing aerosols to the midtroposphere and surface in a climate model and found that the average surface temperature response was opposite in sign to the calculation of the instantaneous or adjusted forcing in their model. Moreover the temperature response of their model was much larger if the absorbing aerosols were added to the middle tropospheric layers rather than the surface layers, a response that was found previously by *Cess et al.* [1985]. We find that our calculated relaxed forcing with longwave forcing included is qualitatively consistent with *Hansen et al.*'s [1997] results. Thus the calculation of relaxed forcing changes the sign of the forcing from that associated only with the instantaneous or adjusted forcing calculation. Moreover, the addition of biomass aerosols to the midtroposphere causes a negative semidirect effect that is larger than that associated with the semidirect effect of fossil fuel soot aerosols which are added mainly to the surface layers in the model. This is shown directly by comparing the relaxed forcing for midtropospheric injection and surface injection of biomass aerosols. Interestingly, whereas the instantaneous forcing is larger (more negative) for surface injection, the relaxed forcing is smaller (less negative) for surface injection of biomass aerosols.

[21] The effect of including the indirect forcing by fossil fuel soot and biomass smoke is to increase the outgoing shortwave radiation associated with the change in the droplet concentration or effective radii in clouds. This leads to a calculation of the overall indirect plus direct shortwave instantaneous forcing of -0.05 Wm^{-2} and -1.21 Wm^{-2} for fossil fuel soot and biomass smoke, respectively. The shortwave relaxed forcing for fossil fuel soot is larger (more positive) than the instantaneous forcing for fossil fuel soot, i.e., 0.12 ± 0.30 Wm^{-2} instead of -0.05 Wm^{-2}. Thus, despite the overall negative shortwave forcing associated with including the indirect effects of these aerosols, when clouds are allowed to decrease as a result of the heating by these aerosols, the result is a net positive, albeit small, shortwave relaxed forcing. The shortwave relaxed forcing associated with biomass aerosols is more negative than is the calculation of instantaneous forcing, i.e., -1.40 ± 0.52 Wm^{-2} instead of -1.21 Wm^{-2}. As in the case with direct effects only, this latter effect is associated with the increase in low-level clouds for biomass smoke (Figure 2b).

[22] The longwave forcing associated with indirect plus direct fossil fuel soot effects is somewhat smaller (less negative) than the fossil fuel direct effects because the temperature change is smaller (Figure 2a). The longwave forcing associated with indirect plus direct forcing by biomass smoke is also somewhat smaller (less negative) than that for direct effects only. In the latter case, the change in precipitable water explains most of this difference (Table 3). The total (shortwave and longwave) relaxed forcing for fossil fuel soot indirect plus direct effects is only 0.01 ± 0.25 Wm^{-2}. Thus these calculations suggest that emissions of fossil fuel soot have almost no effect on climate. The biomass smoke indirect plus direct total relaxed forcing is -1.71 ± 0.44 Wm^{-2}. Thus these aerosols may have important cooling effects which are not fully captured if only their instantaneous forcing is considered. The total semidirect forcing is $+0.06 \pm 0.25$ Wm^{-2} for fossil fuel soot aerosols and -0.50 ± 0.44 Wm^{-2} for biomass smoke.

[23] The decrease in cloudiness in these calculations is related to the increase in the total absorbed shortwave radiation in the atmosphere (Table 4). We evaluated the total absorbed radiation as the difference in the net outgoing shortwave radiation at the top of the atmosphere minus that at the surface between the perturbed case (direct or direct plus indirect) and the preindustrial case. The absorbed shortwave radiation for fossil fuel soot direct effects is 0.95 ± 0.10 Wm^{-2} and is 0.88 ± 0.10 Wm^{-2} for fossil fuel soot indirect plus direct effects. The absorbed shortwave radiation for biomass aerosols is 2.09 ± 0.11 Wm^{-2} and 1.89 ± 0.10 Wm^{-2} for the direct and indirect plus direct cases, respectively. These results are only slightly changed when the instantaneous forcing is considered instead of the relaxed forcing.

[24] When biomass aerosols, fossil fuel soot, and anthropogenic sulfate are all included, we calculate a globally and annually averaged change in stratiform cloud amount of -0.29 ± 0.28 gm^{-2} for midtroposphere injection of biomass smoke or -0.20 ± 0.22 gm^{-2} for surface injection of biomass smoke, respectively, in the case of direct effects only. Our calculated total relaxed forcing for these two cases is -1.24 ± 0.35 and -0.91 ± 0.23 Wm^{-2}, respectively, compared to the instantaneous calculations of -0.90 and -1.01 Wm^{-2}, respectively. The indirect plus direct relaxed forcing associated with these cases is -2.90 ± 0.41 and -1.98 ± 0.32 Wm^{-2} compared to the instantaneous forcing calculations of -2.40 and -1.97 Wm^{-2}. The smaller forcings in the case of surface injection are consistent with the findings for the biomass-only case when smoke aerosols are injected at the surface rather than in the midtroposphere (see Table 2).

Table 4. Annual Average Shortwave Radiation Absorbed in the Atmosphere in the Simulations[a]

Case	Shortwave, W/m^2
Fossil fuel, direct	0.95 (0.10)
Biomass, direct	2.09 (0.11)
Biomass, surface,[b] direct[c]	0.83 (0.10)
Fossil + biomass, direct	3.10 (0.23)
All (fossil + biomass + sulfur), direct	3.02 (0.12)
All, surface,[b] direct	1.72 (0.12)
Fossil fuel, indirect + direct	0.88 (0.10)
Biomass, indirect + direct	1.89 (0.10)
Biomass, surface,[b] indirect + direct[c]	0.77 (0.10)
Fossil + biomass, indirect +direct	2.80 (0.12)
All, indirect + direct	2.73 (0.10)
All, surface,[b] indirect + direct	1.61 (0.08)

[a]Numbers in parentheses are the standard deviations of the 10 (or 9) annual averages.
[b]Biomass aerosols were injected at the surface in these simulations.
[c]These simulations were only 9 years.

4. Conclusions

[25] Previously, it has been suggested that the absorption of radiation by aerosols would lead to reduced cloudiness and an offset of the net cooling mechanisms associated with aerosols [*Ackerman et al.*, 2000; *Hansen et al.*, 2000]. In contrast, our calculation of the relaxed forcing shows that the total (longwave plus shortwave) relaxed forcing is smaller (more negative) than the instantaneous or adjusted forcing. Thus smoke and soot have a diminished warming effect or even a net cooling effect in the atmosphere if the changes in cloudiness and atmospheric temperature associated with the short timescale response to these aerosol types are included in the calculation of forcing. In addition, the effects of absorbing soot and smoke on direct and indirect forcing depend on the altitude of the aerosols. Smoke injected in the middle troposphere causes a larger increase in local temperatures, a decrease in the overall cloudiness in midtroposphere levels, and, as a result, a decrease the longwave TOA relaxed forcing (i.e., a larger negative forcing). An increase in stratiform cloudiness at lower altitudes is associated with a net overall cooling at these levels and results in a negative (or very small positive) shortwave relaxed forcing. These factors substantially decrease the expected positive semidirect effect associated with absorbing aerosol and are consistent with the sign of the surface temperature response calculated for absorbing aerosols placed at different altitudes in the model of *Hansen et al.* [1997].

[26] *Lohmann and Feichter* [2001] previously showed that when the indirect effects of sulfate, fossil fuel soot, and biomass smoke are considered together there is a combined direct plus indirect shortwave relaxed forcing of -1.3 Wm^{-2}

in columns where the BC burden exceeds 2 mgW m^{-2}, but their calculation did not include a self-consistent calculation of the effects of BC absorption when the BC is included in droplets in the indirect effect. Our calculation for the shortwave relaxed forcing in the case of "all" anthropogenic aerosols in regions with BC > 2 mg m^{-2} is -3.0 W m^{-2} and -3.1 W m^{-2} for surface injection of biomass aerosols and for midtropospheric injection, respectively. Differences in this calculation of forcing are especially sensitive to how much of the BC with >2 mg m^{-2} is located over ocean because aerosol scattering has a larger impact over this lower albedo surface. In addition, our direct instantaneous forcing of scattering anthropogenic aerosol components may be more negative than that in the *Lohmann and Feichter* [2001] model since our treatment of grid-averaged relative humidity allows values up to 100% [*Penner et al.*, 1998]. Also, whereas *Lohmann and Feichter* [2001] find little effect from the consideration of longwave forcing, the component of the longwave forcing associated with our simulation for two cases is -0.4 W m^{-2} and -0.8 W m^{-2} for surface injection and midtroposphere injection of biomass aerosols, respectively, in regions where the BC column is greater than 2 mg m^{-2}.

[27] Fossil fuel soot injected near the surface on average warms the atmosphere at all levels and causes a decrease in cloudiness near the surface layer where the soot is injected. The total shortwave and longwave relaxed forcing by these aerosols remains near zero suggesting a near zero climate effect for both the direct and the indirect plus direct effects rather than being strongly positive as by *Jacobson* [2002]. Our calculation of the effects of fossil fuel soot leads to a net forcing that is not significantly different from zero, in contrast to the apparent forcing needed to reproduce the climate model results of *Jacobson* [2002] which requires a positive net forcing of approximately 0.5 Wm^{-2} in a simple climate model that approximates his results [*Penner*, 2003]. While we have no explanation for the difference between our model and that of *Jacobson* [2002], such differences may point to important uncertainties with respect to the effects of soot and smoke. Different treatments of internal versus external forcing by combined BC and OM, as well as the treatment of humidity effects and changes in droplet concentrations will cause differences in the instantaneous and/or relaxed forcing calculated from different models. Moreover, because of differences associated with the altitude of the aerosols, differences in the treatment of convective lofting will cause further differences between models. However, the decreased relaxed forcing associated with including the semidirect effect should be a general conclusion that is valid across all models. We are now in the process of calculating the temperature response from an atmospheric general circulation model that is coupled to a slab ocean model to calculate the long term temperature response to these aerosols as well as the radiative changes associated with the second indirect effect. This will allow us to evaluate whether the relaxed forcing concept can be used with absorbing aerosols to maintain an approximate linear relationship between "forcing" and the global average surface temperature change.

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