

Fossil Fuel and Biomass Burning Effect on Climate—Heating or Cooling?

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ABSTRACT

Emission from burning of fossil fuels and biomass (associated with deforestation) generates a radiative forcing on the atmosphere and a possible climate change. Emitted trace gases heat the atmosphere through their greenhouse effect, while particulates formed from emitted SO_2 cause cooling by increasing cloud albedos through alteration of droplet size distributions. This paper reviews the characteristics of the cooling effect and applies Twomey's theory to check whether the radiative balance favors heating or cooling for the cases of fossil fuel and biomass burning. It is also shown that although coal and oil emit 120 times as many CO_2 molecules as SO_2 molecules, each SO_2 molecule is 50–1100 times more effective in cooling the atmosphere (through the effect of aerosol particles on cloud albedo) than a CO_2 molecule is in heating it. Note that this ratio accounts for the large difference in the aerosol (3–10 days) and CO_2 (7–100 years) lifetimes. It is concluded, that the cooling effect from coal and oil burning may presently range from 0.4 to 8 times the heating effect. Within this large uncertainty, it is presently more likely that fossil fuel burning causes cooling of the atmosphere rather than heating. Biomass burning associated with deforestation, on the other hand, is more likely to cause heating of the atmosphere than cooling since its aerosol cooling effect is only half that from fossil fuel burning and its heating effect is twice as large. Future increases in coal and oil burning, and the resultant increase in concentration of cloud condensation nuclei, may saturate the cooling effect, allowing the heating effect to dominate. For a doubling in the CO_2 concentration due to fossil fuel burning, the cooling effect is expected to be 0.1 to 0.3 of the heating effect.

1. Introduction

The human race is conducting an "experiment" on the sensitivity of the earth-atmosphere system to fast changes in the environment. Fossil fuel use has doubled in the last 20 years (Rotty 1987), and every year hundreds of thousands of man-made fires are destroying the tropical forest (Crutzen et al. 1979; Seiler and Crutzen 1980; Malingreau and Tucker 1988; Kaufman et al. 1990; Setzer 1988; Setzer and Pereira 1989; Tucker et al. 1984), filling millions of square kilometers with dense smoke (Andreae et al. 1988). Carbon dioxide concentration has increased 3% in the last decade (Houghton and Woodwell 1989), sulfate concentration in Greenland ice has tripled in the last century (Mayewski et al. 1986), and the particle concentration has increased over most of the northeastern United States, increasing the solar extinction coefficient by 50% during summer months (Husar et al. 1978). This "experiment" may change the radiative balance of the earth, and there is a potential danger that it may adversely

impact our climate. Yet, due to uncertainty in the aerosol effects (as well as other uncertainties, Schneider 1989), the direction to which the "experiment" is taking us and its possible twists are yet to be established (Wigley 1989).

As a result, it is important to review the relative effects of the gases and particles emitted from fossil fuel and deforestation-related biomass burning. Climate models, which have been used in simulating the climate change, have accounted for the effect of CO_2 and other trace gas emissions (Hansen et al. 1984; Hansen and Lebedeff 1987), but the complex effect of particles on the radiative budget, through alteration of cloud albedo (Twomey 1977a), has only recently been introduced into the climate models (Wigley 1989).

Particles formed in the air from SO_2 emitted by fossil fuel burning (or organic particles emitted directly from biomass burning) may affect the radiation budget and boundary layer meteorology by directly reflecting and absorbing incident sunlight (Coakley et al. 1983; Shaw 1983). The radiation budget and boundary layer meteorology may be affected even more so by increasing the number of available cloud condensation nuclei (CCN), thus altering cloud droplet distributions and affecting cloud albedos (Twomey et al. 1984). These anthropogenic particles are a major source of cloud

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condensation nuclei (Twomey 1977a; Charlson et al. 1987). An increase in the aerosol concentration may cause an increase in the reflectance of thin to moderate clouds (Twomey 1977a; Coakley et al. 1987) and a decrease in the reflectance of thick clouds mainly due to absorption by soot (Twomey 1977a). Since for most clouds the aerosol effect dominates through an increase in the cloud albedo, Twomey et al. (1984) suggested that the net cooling effect due to an increase in the aerosol concentration can be as strong as the heating effect due to the global increase of CO₂ and other trace gases. The effect of soot on cloud albedo may be significant in urban areas. However, no significant aerosol absorption was found in summer pollution over the eastern United States or over tropical regions contaminated by smoke from biomass burning associated with deforestation (Kaufman et al. 1990; Setzer 1988; Setzer and Pereira 1989) where soot concentration was measured to be 7% of the aerosol mass (Andreae et al. 1988).

This paper reviews the characteristics of and evidence for the SO₂ induced cooling effect. The basic theory of the effect of pollution on cloud microphysics and its global implications, as suggested by Twomey (Twomey 1977a; Twomey et al. 1984), is applied to compare the relative effect of a small increase in the consumption rate of coal, oil, or biomass burning on heating and cooling of the atmosphere. This perturbation analysis approach allows linearization of the relations, thus simplifying the analysis and reducing the number of uncertain parameters. For biomass burning we restrict the analysis to burning associated with deforestation. Burning of savanna and other organic wastes do not result in a net CO₂ emission and the amount of graphitic carbon emitted is unknown. The cooling effect is reviewed in section 2. Climatic effects of CO₂ and SO₂ are discussed in section 3. The analysis is applied to coal, oil, and biomass burning in section 4. Section 5 concludes this paper.

2. Cooling resulting from particulates

In order to judge the reality of the cooling effect due to particulates, it is necessary to review its characteristics and the evidence presented in the literature for, namely, the effect of particulate pollution on cloud characteristics. Empirical observations of the dependence of cloud droplet concentration on the air mass origin were first reported by Squires (1956). Squires and Twomey (1960) demonstrated that maritime and continental clouds showed considerable difference in their droplet population and in the consequent rainfall. Junge and McLaren (1970) also measured the relation between aerosol size spectra and the resultant CCN concentration. Later, Twomey and Warner (1967) measured the relation between CCN and cloud droplet spectra. They found good agreement with theoretical computations of cloud formation. A direct relation be-

tween microphysical properties of clouds and the aerosol concentration was measured by Hobbs and Rangno (1985) and Radke (1989). They found in comparison of continental and maritime cloud observations that the latter formed in air masses containing small aerosol concentrations and had wide drop radii dispersion (larger radii). Continental clouds were observed to have higher aerosol concentrations, narrower, smaller drop radii distributions, and fewer precipitation events than for maritime clouds (Hobbs and Rangno 1985). Smoke from biomass burning produced a large increase in the CCN concentration for a maritime air mass, even though the aerosol particles were of organic origin (Squires and Twomey 1960; Hobbs and Radke 1969).

A valid question is whether an increase in aerosol concentration can affect clouds that are already rich in CCN. Braham (1974) studied the effect of urban pollution on low stratus clouds and found a roughly proportional increase in CCN from 1000 cm⁻³ in clean, rural areas to 3000 cm⁻³ in polluted regions. Leitch et al. (1986) investigated the relation between aerosol concentration, CCN, and cloud droplet concentration (N_d) in regions with stratiform clouds and moderately convective clouds. They also found that both N_d and CCN increased with particle concentrations if CCN was less than 1500 cm⁻³ and that N_d became saturated at around 1000 cm⁻³. Egan et al. (1974) found that even for a background N_d of 1000 cm⁻³, an increased aerosol concentration resulted in an additional increase in N_d up to 3000 cm⁻³ and a decrease in the drop size radii from 12 to 8.5 μm. Therefore, for N_d concentrations less than 1000 cm⁻³, there is a direct relation between the presence of particulate pollution, CCN, and N_d . Above this value the effect of an increase in aerosol concentration on the cloud characteristics may saturate; the value of N_d for which saturation occurs is uncertain however.

Coakley et al. (1987) measured the effect of particulate pollution on cloud albedo. They noticed that stratocumulus clouds increased in brightness in the visible and 3.7 μm channels above ship tracks, roughly in agreement with Twomey's theory (1977a). A large increase in the visible reflectance was also reported by Conover (1966) for stratocumulus clouds affected by ship tracks. On the other hand, Schwartz (1988) in discussion of the possible effects of oceanic dimethyl sulfide (DMS) on cloud albedo, pointed out that if anthropogenic pollution, which is greater in the Northern Hemisphere than in the Southern Hemisphere, has a strong effect on cloud albedo and climate, there should be a difference between the two hemispheres in the cloud albedo and in the temperature record. He did not find a difference. However, the Northern Hemisphere may be cooling relative to the Southern Hemisphere (Wigley 1989) if the temperature records are biased due to the heat-island effect of small cities on temperature measurements (Karl and Jones 1989).

TABLE 1. Production of CO₂ and SO₂.

Burning substance	CO ₂ [10 ¹⁵ (gC/year)]	SO ₂ [10 ¹⁵ (gS/year)]
Coal in 1984	2.1 (Rotty 1987)	0.05
Oil in 1984	2.2 (Rotty 1987)	0.05
Gas in 1984	0.8 (Rotty 1987)	0.00
Total fossil fuel	5.6 (Houghton and Woodwell 1989)	0.13
Biomass burning	2.7 (Crutzen et al 1985)	~0 (McMahon 1983)
Biomass burning	2-5 (Houghton and Woodwell 1989)	

3. Climatic effects of SO₂ and CO₂

Three burning processes (coal, oil, and deforestation related biomass) are examined, since they represent most of the anthropogenic effect on climate (other effects include emissions of chlorofluorocarbons, emission of CH₄ from agricultural practices, and emissions related to burning of agricultural wastes and savannas). Fossil fuel contributes 75% of the anthropogenic SO₂ emitted to the atmosphere and 40% of total sulfur emission (Schwartz 1988). Coal, oil, and deforestation related biomass burning each contribute around one-third of the global source of anthropogenic CO₂. In Table 1 the emission rates of these processes are summarized. We first examine the climatic effect of CO₂ and SO₂.

a. Heating by CO₂

According to the model of Hansen et al. (1984) and Hansen and Lebedeff (1987), doubling of CO₂ (corresponding to a change in CO₂ concentration of $\Delta C = 320$ ppm) causes a change in the energy balance due to the greenhouse effect, which is equivalent to an increase in the solar constant (F_0) of $\Delta F_0/F_0 = 2\%$. The relation between the CO₂ concentration and the radiative forcing due to CO₂ infrared emission is nonlinear due to the band structure of the CO₂ absorption. Using the model of Peng et al. (1987) a numerical relation was computed between the infrared flux emitted by CO₂ and the CO₂ concentration (Chou, private communication). This relation can be approximated by

$$\frac{\Delta F_0}{F_0} = 1.6 \times 10^{-4} \Delta C^{0.84} \quad (1)$$

where the coefficient was chosen to fit a change of $\Delta F_0/F_0 = 0.02$ for $\Delta C = 320$ ppm. The relation between an increment in the CO₂ rate of emission, R_c (in units of molecule per cm² day⁻¹), and the resulting increase in the CO₂ concentration, ΔC (in ppm), is

$$R_c [\text{cm}^{-2} \text{ day}^{-1}] = \Delta C \times 10^{-6} \frac{AH}{365V_m T_c} \quad (2)$$

where A is Avogadro number and V_m is the volume of one mole (22 400 cm³). This relation is based on an exponential vertical distribution of CO₂ with a scale height of H (see Table 2) and an exponential response to a change in the atmospheric concentration of CO₂ with an effective lifetime T_c (see section 3d). Note that in this and the following equations T_c is defined in years and aerosol lifetime T_a in days. Substituting values for A , V_m , and H one gets

$$R_c [\text{cm}^{-2} \text{ day}^{-1}] = 5.9 \times 10^{16} \frac{\Delta C}{T_c} \quad (3)$$

From Eqs. (1) and (3):

$$\frac{\Delta F_0}{F_0} = 1.3 \times 10^{-18} (R_c T_c)^{0.84} \quad (4)$$

For a change in the concentration of CO₂ of $\Delta C = 50$ ppm (from a base value of 320 ppm), the relation between $\Delta F_0/F_0$ and ΔC can be approximated by a linear relation:

$$\frac{\Delta F_0}{F_0} = 1.5 \times 10^{-21} R_c T_c \quad (4')$$

The heating rate h_c (relative to the solar flux F_0) due to an increase in CO₂ emission rate of one unit (one molecule per cm² day⁻¹) is

TABLE 2. Parameters used in Eqs. (1) and (2).

Parameter	Value	Source
f_c	Fraction of carbon	0.8 (Hidy 1974)
f'_s	Fraction of sulfur	0.02 ± 0.005 (Wixson et al. 1980)
e_s	Emission efficiency	90% (Semb 1978)
$f_s = f'_s \cdot e_s$	Fraction of emitted sulfur	0.018 ± 0.005
f_s	Fraction of emitted sulfur	0.019 (Seinfeld 1986)
H	Scale height of CO ₂	8 km

$$h = \frac{\Delta F_0}{F_0} / R_c = 1.5 \times 10^{-21} T_c. \quad (5)$$

b. Cooling by SO₂

In order to compare the effects of SO₂ and CO₂ on the radiative balance, the cooling rate due to particulates from emitted SO₂ is computed. The computation of the SO₂ cooling rate involves relationships of the following quantities:

- SO₂ emission rate increase R_s [molecules per cm² day⁻¹] and the aerosol concentration N_a ;
- N_a and the cloud condensation nuclei concentration N_c ;
- N_c and the corresponding cloud droplet concentration N_d ;
- N_d and the cloud optical thickness τ_c ;
- τ_c and the cloud albedo A_c ; and
- a change in the cloud albedo ΔA_c and a corresponding change in the solar constant $\Delta F_0/F_0$.

The derivation does not include the effect of soot (since its concentrations are usually very small), possible increases in cloud lifetimes (Hobbs and Rangno 1985; Radke et al. 1990) or cloud fraction (in very clear areas), since these effects were not studied enough to be quantified. Low and midlevel clouds are essentially black in the infrared (Twomey et al. 1984). Therefore, it is also assumed that clouds modified by pollution have the same thermal characteristics as unmodified clouds. In the following equations, the relations that comprise the cooling effect are developed.

1) RELATION BETWEEN THE SULFUR RATE OF EMISSION AND THE CCN

An increase in the rate of coal burning generates an increase of R_c [cm⁻² day⁻¹] in the CO₂ emission rate and an accompanying increase of R_s [cm⁻² day⁻¹] in the SO₂ emission rate. Since industrial pollution covers only a fraction of the earth surface (α), while CO₂ is uniformly distributed, the increase in concentration (ΔN_s) of sulfur atoms, for a scale height of 2 km is

$$\Delta N_s [\text{cm}^{-3}] = 5 \times 10^{-6} R_s T_a / \alpha \quad (6)$$

where T_a is the lifetime of an aerosol particle in the atmosphere. Garland (1978) suggested that the lifetime of an aerosol particle that originated from SO₂ emission is $T_a = 5$ days. The corresponding extra sulfur mass (Δm_s) is

$$\Delta m_s [\text{g cm}^{-3}] = \Delta N_s \frac{M_s}{A} \quad (7)$$

where M_s is the atomic weight of sulfur ($M_s = 32$) and the aerosol mass (Δm_a):

$$\Delta m_a [\text{g cm}^{-3}] = \Delta m_s \frac{f_{sp}}{f_{ws}} \quad (8)$$

where f_{sp} is the estimated fraction of SO₂ converted to sulfate particles $f_{sp} = 0.4 \pm 0.1$ (Seinfeld 1986; Georgii 1979; Husar 1978; Rodhe 1978) and f_{ws} is the fraction of the particle's weight that is composed of sulfur ($f_{ws} = 0.12$) (Fraser et al. 1984).

It is possible that not all of the sulfate aerosol particles participate as CCNs. The fraction of the aerosol mass that participate in cloud formation depends on the interaction of potential CCNs and the water vapor in a cloud. Pruppacher and Klett (1980) show that adjustment of a particle to a new humidity is nearly instantaneous; therefore the rate in which the maximum supersaturation is reached does not affect the fraction of aerosol particles that participate in cloud formation. They also computed the effect of aerosol particles on cloud formation with a model of maritime aerosol and a model of continental aerosol using an entrainment model. This simulation showed a strong relation between particle concentration and cloud drop concentration. Similar strong correlations between aerosol particles, CCNs, and cloud drop concentration were measured by Leitch et al. (1986) up to a maximum cloud drop concentration of $N_d = 1000$.

The relation between CCN concentration and particle concentration can be derived from the theoretical relation between the critical saturation and the particle radius (Twomey 1977b):

$$r_{min} [\mu\text{m}] = 0.0145 S^{-2/3} \quad \text{for } S = 0.1-0.5 \rightarrow r_{min} = 0.02-0.07 [\mu\text{m}]. \quad (9)$$

Here, r_{min} is the minimum particle radius for a given supersaturation S [%] that can form a cloud droplet. Aerosol particles that are formed from airborne SO₂, are in the accumulation mode, and can be described by a lognormal distribution with a number distribution median radius $r_g = 0.05 \mu\text{m}$ and standard deviation of the logarithm of the radius of $\sigma = 0.6$ (Whitby 1978). The aerosol mass is therefore concentrated around a mass distribution mean radius of $r_m = 0.15 \mu\text{m}$ [for a lognormal distribution $r_m = r_g \exp(3\sigma^2)$]. Since 85% of the mass occurs for $r > 0.07 \mu\text{m}$, most of the sulfate aerosol mass can participate in the cloud formation.

It is possible that some fraction of the sulfate aerosol particles do not participate in cloud formation, due to inhomogeneity in the supersaturation. Inhomogeneous convection and entrainment of dry air can cause such inhomogeneity. Therefore, it is appropriate to assume that some fraction f_{ac} of the aerosol mass Δm_a is the actual mass of the CCNs (Δm_c):

$$\Delta m_c [\text{g} \cdot \text{cm}^{-3}] = \Delta m_a \cdot f_{ac} \quad (10)$$

where f_{ac} determines the fraction of aerosol mass that participate in cloud formation. It is assumed that f_{ac} ranges between 0.25 to 0.75 based on the measurements of Leitch et al. (1986—Fig. 3).

The number density ΔN_c associated with that mass is

$$\Delta N_c [\text{cm}^{-3}] = \Delta m_c / (1.5 V_a) \quad (11)$$

where V_a is the particle volume for radius r_m [μm]. A density of $1.5 [\text{g cm}^{-3}]$ is used. Combining Eqs. (6)–(8), (10), and (11) one gets

$$\Delta N_c [\text{cm}^{-3}] = 1.4 \times 10^{-16} R_s \frac{T_{ac} f_{ac}}{\alpha r_m^3} \quad (12)$$

2) RELATION BETWEEN CLOUD DROP CONCENTRATION AND CCN

The relation between the cloud drop concentration (N_d) and the CCN concentration (N_c) is typically (Twomey 1977b; Twomey 1959):

$$N_d \propto N_c^{2/(k+2)} \quad (13)$$

where k was found empirically as $k \approx 0.5$ (Twomey 1977b). The value of k is important, since for higher values of k , the cloud drop concentration N_d is less dependent on variations of the CCN concentration caused by an increase in pollution levels. Therefore, a higher value of k for a higher aerosol concentration can be a stabilizing mechanism, which could make cloud characteristics insensitive to increases in pollution levels in an already polluted environment. The relation between the cloud drop concentration and the CCN takes into account the effect of CCN concentration on the maximum supersaturation and was used successfully to predict cloud drop concentration (Twomey and Warner 1967). The value of k in Eq. (13) is based on the relation between the CCN and the supersaturation S (where S is the supersaturation above

100% relative humidity, Twomey 1959). In order to assess the value and uncertainty in the parameter k , some published measurements of k and the CCN concentrations are given in Table 3. Most of the values are in the 0.5–1.0 range. The average value of k for continental air measured by Braham (1974) and given in Table 3 is $k = 0.8 \pm 0.15$. The difference between these two values of k results in a small uncertainty of 10% in the droplet concentration.

For a small change ΔN_c the corresponding change in N_d (ΔN_d) from Eq. (13) is

$$\frac{\Delta N_d}{N_d} = \left(\frac{2}{k+2} \right) \frac{\Delta N_c}{N_c} = 0.7 \frac{\Delta N_c}{N_c} \quad (14)$$

Presently, the global average CCN concentration is estimated at 125 CCN cm^{-3} (Twomey et al. 1984). Since pollution affects clouds residing mainly over land and nearby ocean areas, where the sources of anthropogenic CCN production are located, and where the CCN concentrations are larger (see Table 3), a higher average value should be used. The average CCN concentration in Table 3 for maritime and continental air masses for a super saturation of 0.3% is used ($N_c = 300 \pm 200$).

3) RELATION BETWEEN N_d , τ_c AND A_c

The increase in N_d causes an increase in cloud optical thickness (τ_c) of $\Delta \tau_c$. Assuming a fixed amount of liquid water and scattering based on geometrical optics by the cloud droplets (Twomey et al. 1984):

$$\frac{\tau_c + \Delta \tau_c}{\tau_c} = \left(\frac{N_d + \Delta N_d}{N_d} \right)^{1/3} \quad (15)$$

TABLE 3. Measurements of k and the CCN concentration in several air masses at two supersaturations S .

k	CCN (cm^{-3})		Air mass	Source
	1% S	0.3% S		
0.7	100	40	Maritime	(Twomey and Wojciechowski 1969)
0.5	600	400	Continental	(Twomey and Wojciechowski 1969)
0.4	2000	1200	Continental	(Twomey 1959)
0.3	125	90	Maritime	(Twomey 1959)
0.9	3500	1200	Buffalo	(Kocmond 1965)
0.46	53	30	Hilo	(Jiusto 1967)
0.46	105	60	Hawaii	(Jiusto 1967)
0.74	82	30	Ocean	(Radke 1989)
0.76	599	240	Coast	(Radke 1989)
1.34	569	110	90 km in land	(Radke 1989)
3.45	892	14	185 km in land	(Radke 1989)
0.69	2210	960	Up wind St. Louis	(Braham 1974) July, August 1971
0.75	3890	1600	Down wind St. Louis	(Braham 1974) July, August 1971
0.83	848	300	Up wind St. Louis	(Braham 1974) Feb, March 1971
0.97	1678	500	Down wind St. Louis	(Braham 1974) Feb, March 1971
0.63	1166	500	Up wind St. Louis	(Braham 1974) July, August 1972
0.65	1625	700	Down wind St. Louis	(Braham 1974) July, August 1972
0.96	977	300	Up wind St. Louis	(Braham 1974) March, April 1973
1.08	1451	400	Down wind St. Louis	(Braham 1974) March, April 1973
		50 ± 20	Average maritime	
		600 ± 400	Average continental	
		900 ± 450	Average urban	

and for a small change $\Delta\tau_c$:

$$\frac{\Delta\tau_c}{\tau_c} = 0.33 \frac{\Delta N_d}{N_d}. \quad (16)$$

The relation between the cloud albedo (A_c) and the cloud optical thickness (τ_c) is (Twomey et al. 1984):

$$\Delta A_c = \gamma(\Delta\tau_c/\tau_c) \quad \text{where } \gamma = 0.15\text{--}0.20$$

for $0.3 < A_c < 0.9$. (17)

Combining Eqs. (12), (14)–(17) one gets

$$\Delta A_c = 6 \times 10^{-18} R_s \left(\frac{T_a f_{ac}}{N_c r_m^3 \alpha} \right). \quad (18)$$

4) RELATION BETWEEN ΔA_c AND THE RADIATIVE BALANCE

Assuming that a fraction β of each unit of polluted area is covered by clouds that can be affected by the pollution (Charlson et al. 1987; Schwartz 1988), and that the emitted pollution covers a fraction α of the earth's surface, the change in radiative balance that results from a change in cloud albedo ΔA_c is equivalent to a change in the solar constant ($\Delta F_0/F_0$) of

$$\frac{\Delta F_0}{F_0} = \Delta A_c \frac{\alpha\beta}{1-a}. \quad (19)$$

The factor $(1-a)$ was introduced, where a is the total albedo of the earth and its atmosphere ($a = 0.3$) (Barkstrom et al. 1989), since only $(1-a)$ fraction of the solar flux is absorbed by the earth surface and the atmosphere. It is estimated that the value of β ranges between 0.2 and 0.5. From Eqs. (18) and (19):

$$\frac{\Delta F_0}{F_0} = 8 \times 10^{-18} R_s \frac{T_a f_{ac} \beta}{N_c r_m^3}. \quad (20)$$

Note that as long as the radiative effect is proportional to R_s (as is the case for a perturbation model), α is canceled in Eq. (20), since a smaller surface cover would correspond to a higher aerosol rate of emission for the same total emission. The cooling rate h_s (relative to the solar flux) due to an increase in SO_2 emission rate of one unit (one molecule per $\text{cm}^2 \text{ day}^{-1}$) is

$$h_s = \frac{\Delta F_0}{F_0} / R_s = 8 \times 10^{-18} \frac{T_a f_{ac} \beta}{N_c r_m^3}. \quad (21)$$

c. Comparison between CO_2 and SO_2

The ratio (η) between cooling induced by SO_2 and heating by CO_2 for a unit rate of emission for each molecule is given by

$$\eta = \frac{h_s}{h_c} = \frac{(\Delta F_0/F_0)/R_s}{1.5 \times 10^{-21} T_c} \quad (22)$$

where h_c and h_s were substituted from Eqs. (5) and (21). For the linear solution, using Eq. (21) we get

$$\eta = \frac{8 \times 10^{-18} T_a f_{ac} \beta / (N_c r_m^3)}{1.5 \times 10^{-21} T_c} = 5300 \frac{T_a f_{ac} \beta}{T_c N_c r_m^3}. \quad (23)$$

Although the linearization used in Eq. (14) and (16) served us well by allowing cancellation of the fraction α , which defines the fraction of the globe that is covered by pollution (and is difficult to quantify), it may be interesting to consider the full solution in order to assess the impact of a large change in the emission (e.g., corresponding to doubling of CO_2). Following the analysis that led to Eq. (21), but without the linearization in Eqs. (14) and (16), the relation between the increase in the rate of emission of $\text{SO}_2 - R_s$ and the equivalent change in the solar constant $\Delta F_0/F_0$ due to the resulting cooling is computed by substituting Eqs. (12), (13), (15), and (17) into Eq. (19):

$$\frac{\Delta F_0}{F_0} = \frac{\alpha\beta\gamma}{1-a} \times \left[\left(1 + \frac{1.4 \times 10^{-16} R_s T_a f_{ac}}{N_{co} \alpha r_m^3} \right)^{2/3(k+2)} - 1 \right]. \quad (24)$$

This expression is solved for the increase in the emission rate of SO_2 (R_s), required to generate a radiative change of $\Delta F_0/F_0$:

$$R_s = \frac{N_{co} \alpha r_m^3}{1.4 \times 10^{-16} T_a f_{ac}} \times \left\{ \left[\frac{\Delta F_0}{F_0} \frac{(1-a)}{\alpha\beta\gamma} + 1 \right]^{1.5(k+2)} - 1 \right\}. \quad (25)$$

The ratio between cooling and heating due to consumption of a unit mass of fossil fuel, given by Eq. (23), can be written for the nonlinear solution by substituting R_s from Eq. (25) for $k = 0.8$ and $\gamma = 0.17$ [see Eq. (17)] into Eq. (22):

$$\eta = 10^5 \frac{(\Delta F_0/F_0) f_{ac} T_a}{T_c N_c r_m^3 \alpha} \left[\left(1 + \frac{4\Delta F_0/F_0}{\alpha\beta} \right)^{4.2} - 1 \right]^{-1}. \quad (26)$$

Equation (26) does not account for nonlinearity in the CO_2 radiative heating [Eq. (1)] or in the relation between cloud albedo and cloud optical thickness [Eq. (17)]. These two nonlinear effects are much smaller than the nonlinearity in the relation between cloud optical thickness and aerosol concentration. Doubling of the CO_2 concentration generates an error of 20% in the linearization of Eq. (4). The nonlinearity in the relation between ΔA_c and $\Delta\tau_c/\tau_c$ [Eq. (17)] can generate an error of 10%–20% for a large change of ΔA_c of 0.2–0.4. The nonlinearity in the relation between the cloud optical thickness and the aerosol concentration can decrease the ratio between SO_2 cooling and CO_2 heating [described later by Eq. (30) and Table 7] from 1.2 for a small change in the consumption of SO_2 to 0.2 for a doubling in the CO_2 concentration, if the pollution covers 20% of the earth surface ($\alpha = 0.2$).

d. Carbon dioxide lifetime

Siegenthaler and Oeschger (1978) showed that the response of atmospheric CO₂ for a delta-function input is not exponential. Using a box diffusion model, they computed a relationship between the excess amount of CO₂ and the time passed from the injection of one unit of CO₂. A fast response occurs during the first two years, due to transfer of part of the excess CO₂ from the atmosphere to the oceanic mixing layer, followed by a slower diffusion of the CO₂ to the deeper ocean. Therefore, the effective lifetime of excess CO₂ depends on the time scale of the climatic effect.

If we compare the heating and cooling effects due to an excess burning of one unit per year of coal, oil, or biomass during the next Y years, then the average lifetime (\bar{T}) of the excess CO₂ produced in the burning process is found from a double integral of the response function $f_i(t)$ (Siegenthaler and Oeschger 1978):

$$\bar{T}(t) = \int_0^Y \frac{dt}{Y} \int_t^Y f_i(t-t') dt' \quad (27)$$

where the external integral is over the emission period of excess CO₂, while the internal integral is over the response to the excess concentration $f_i(t)$ given by Siegenthaler and Oeschger (1978). The average lifetimes \bar{T} computed by Eq. (24) for three values of Y are listed in Table 4. Note that \bar{T} is smaller than the effective lifetime for an impulse emission of CO₂. A similar exponential series for the decay of an impulse of CO₂ was computed (Maier-Reimer and Hasselmann 1987) using an oceanic GCM model, and was used later to model the effect of CO₂ on climate (Harvey 1989). The application of this model to the effective CO₂ lifetime as defined in Eq. (24) is also shown in Table 4. A lower boundary of the CO₂ lifetime is the CO₂ turnover time in the atmosphere and is given in Table 5 as $T = 7 \pm 3$ years. These three models suggest that the appropriate choice for the CO₂ lifetime is in the range 7–100 years, with most probable range of 15–30 years.

4. Application to coal, oil, and biomass burning

a. Production of CO₂ and SO₂

The ratio between the number of carbon atoms n_c in a unit mass of coal and the number of sulfur atoms n_s released during burning is given by

TABLE 4. The effective CO₂ lifetime estimated from the results of a box diffusion model (Siegenthaler and Oeschger 1978) and an oceanic GCM (Maier-Reimer and Hasselmann 1987).

Period of excess emission (Y : years)	Average lifetime T (years)	
	Box diffusion model	Oceanic GCM
50	16	18
100	27	32
1000	135	150

TABLE 5. Range of atmospheric CO₂ residence times.

Time (years)	Reference
2–6	(Machta 1972)
≈5	(WMO 1982)
4	(Delwiche and Likens 1977)
2	(Ramanathan et al. 1985)
7	(Bolin 1986)
8.5 ± 2.5	(Bolin 1986)
1.5	(Hay and Southam 1984)
15	(Junge 1972)*
7.7	(Siegenthaler and Oeschger 1978)

* Junge did not include atmosphere–ocean exchanges. An average value of 7 ± 3 years may be computed from these values. The two most extreme values have been excluded from the average.

$$\frac{n_c}{n_s} = \frac{f_c/M_c}{f_s/M_s} = 120 \pm 60 \quad (28)$$

where M_c and M_s are the atomic weights of carbon ($M_c = 12$) and sulfur ($M_s = 32$), and f_c and f_s are the fractions of carbon and sulfur mass emitted in the burning process. Here f_s is the product of the fraction of sulfur in coal (f'_s) and an emission efficiency (e_s). Table 2 summarizes these parameters. Since most of the carbon is released as CO₂ and most of the sulfur is released as SO₂, the ratio in Eq. (28) holds also for numbers of CO₂ and SO₂ molecules. The ratio between cooling and heating (ξ) due to the burning of coal is obtained as the ratio between the function η [Eq. (22)], which represents the ratio between cooling and heating of one SO₂ molecule and one CO₂ molecule] to the ratio of the production rate of CO₂ and SO₂ [Eq. (28)].

Here

$$\xi = \frac{\eta}{120} = 50 \frac{T_a f_{ac} \beta}{T_c N_c r_m^3} \quad (29)$$

and for the nonlinear model [Eqs. (26) and (28)]:

$$\xi = \frac{\eta}{120} = 840 \frac{(\Delta F_0/F_0) f_{ac} T_a}{T_c N_c r_m^3 \alpha} \times \left[\left(1 + \frac{4\Delta F_0/F_0}{\alpha\beta} \right)^{4.2} - 1 \right]^{-1} \quad (30)$$

b. Climatic effects of coal

The ratio (ξ) of the indirect cooling effect of SO₂ and the heating effect of CO₂ for emissions from coal burning is computed for several values of the following parameters:

- the SO₂ lifetime T_a [days];
- the CO₂ effective lifetime T_c [years];
- the fraction of a given unit area of the surface covered by low clouds that can be affected by pollution (β);
- the pre-perturbation CCN concentration N_c [cm⁻³];

- the particle mass distribution mean radius r_m [μm]; and
- the fraction of the particles that participate in the cloud condensation f_{ac} .

The results for the linear model [Eq. (29)] are presented in Table 6. The ratio of cooling to heating at the present level of emission ranges between 0.1 (much more heating) and 300 (overwhelming cooling), but most probably in the 0.4–8 range (as indicated by the outlined ratios in Table 6). Due to the large uncertainty in these results, it is impossible to determine at this stage if the net effect of coal burning is heating or cooling the atmosphere, although the present analysis shows that cooling is more likely.

For a large change in coal consumption, represented by a larger $\Delta F_0/F_0$, Eq. (30) represents the climatic effect. The ratio ξ between cooling and heating is tabulated in Table 7 for several values of the fraction of the earth covered by pollution (α) and several magnitudes of the radiative effect given by $\Delta F_0/F_0$. For $\Delta F_0/F_0 = 0.02$, which corresponds to doubling of CO_2 , the number of CCNs become so large that the cooling effect is saturated and heating is 2 to 16 times larger than cooling (for $\alpha = 0.5$ and 0.1, respectively).

c. Climatic effects of oil

The climatic effects of oil are similar to the effects of coal. The carbon content of oil is $f_c = 0.84$ (Campbell 1986), similar to $f_c = 0.80$ for coal; f_s ranges from 0.5% to 3.5% for oil (Campbell 1986). From detailed data on sulfur emission from oil burning (Semb 1978), on average, the emitted sulfur is $f_s = 1.8\%$ of the mass of fuel oil. Since for fuel oil, probably all emitted sulfur is emitted as SO_2 , the value for f_s is similar to that for coal. Therefore, the ratio of the cooling effect to the heating effect for oil burning is very similar to that for coal and is represented by Tables 6 and 7.

d. Climatic effect of biomass burning

For biomass burning associated with deforestation a similar analysis to that of coal is applied. The ratio

TABLE 7. The ratio ξ for several values of α and several magnitudes of the radiative effects given by $\Delta F_0/F_0$ (where $\Delta F_0/F_0 = 0.02$ corresponds to doubling of CO_2), showing the decrease in the value of ξ with increasing magnitude of the radiative effect. ($N_c = 300 \text{ cm}^{-3}$, $T_a = 5 \text{ d}$, $T_c = 30 \text{ yr}$, $f_{ac} = 0.5$, $\beta = 0.3$, $r_m = 0.15 \mu\text{m}$).

α	$\Delta F_0/F_0$					
	0.0001	0.001	0.005	0.01	0.02	0.05
1	1.2	1.2	1.1	1.0	0.8	0.5
0.5	1.2	1.2	1.0	0.8	0.5	0.2
0.2	1.2	1.1	0.7	0.5	0.2	0.03
0.1	1.2	1.0	0.5	0.2	0.06	0.01
0.05	1.2	0.8	0.2	0.06	0.00	0.00
0.02	1.1	0.5	0.04	0.01	0.00	0.00
0.01	1.0	0.2	0.01	0.00	0.00	0.00

of concentration of organic particles generated in biomass burning to CO_2 concentration is given from the measurements of Ward (Ward 1986; Ward and Hardy 1984):

$$\frac{\text{mass of CO}_2}{\text{mass of particles}} \approx 100. \tag{31}$$

Smoke from biomass burning probably does not include any substantial amount of sulfur [since forest fuel contains between 0.01% to 0.4% sulfur (McMahon 1984)]. For comparison, in the case of coal 120 atoms of carbon are produced for each atom of sulfur. The ratio of the masses of CO_2 (m_{CO}) and aerosol (m_a) produced by coal burning is

$$\frac{m_{\text{CO}}}{m_a} \left[\frac{g_{\text{CO}_2}}{g_{\text{aerosol}}} \right] = 120 \left(\frac{M_{\text{CO}}}{M_s} \right) \frac{f_{\text{ws}}}{f_{\text{sp}}} = 50 \tag{32}$$

where M_{CO} is the molecular weight of CO_2 . Therefore, from Eqs. (31) and (32) the production of aerosol mass relative to CO_2 mass for biomass burning is half of that for coal. The other trace gases (e.g., CH_4 , etc.) produced during biomass combustion can double the greenhouse effect (Ramanathan et al. 1985). Therefore, the ratio of cooling to heating by biomass burning is 0.25 of that for coal. Comparison with the scenarios listed in Tables

TABLE 6. Ratio between the indirect SO_2 cooling and the CO_2 heating (ξ) for a range of parameters (T_s , T_c , f_{ac} , f , N_c , r_m). Bold numerals represent major cooling and *italic* numerals represent major heating. The outlined ratios indicate the most probable values of the parameters.

β	N_c (cm^{-3})	r_m (μm)	f_{ac}	$\frac{N_c r_m^3}{\beta f_{ac}}$	$T_a \text{ (d)}/T_c \text{ (yr)}$							
					0.03	0.05	0.10	0.16	0.30	0.40	0.70	1.50
0.5	100	0.1	0.75	0.27	5.6	9.2	19	30	55	74	130	278
0.3	100	0.1	0.5	0.67	2.2	3.7	7.5	12	22	30	52	112
0.3	300	0.1	0.5	2.0	<i>0.75</i>	1.2	2.5	4.0	7.5	10	17	37
0.3	300	0.15	0.5	6.7	<i>0.2</i>	0.4	0.8	1.2	2.2	3.0	5.2	11
0.2	300	0.15	0.5	10	<i>0.1</i>	0.2	0.5	0.8	1.5	2.0	3.5	7.5
0.2	300	0.15	0.25	20	<i>0.1</i>	<i>0.1</i>	<i>0.2</i>	<i>0.4</i>	0.8	1.1	1.8	3.7
0.2	500	0.15	0.25	34	<i>0</i>	<i>0.1</i>	<i>0.1</i>	<i>0.2</i>	0.4	0.6	1.0	2.2

TABLE 8. List of symbols.

a	total albedo of the earth and its atmosphere
A	Avogadro number
A_c	cloud albedo
ΔC	change in CO ₂ concentration [ppm]
e_s	emission efficiency
F_0	solar constant
f_{ac}	fraction of aerosol particles that participate in cloud condensation
f_c	fraction of carbon mass emitted from fuel
f_s	fraction of sulfur mass emitted from fuel
f'_s	sulfur content of fuel
f_{sp}	fraction of SO ₂ converted to sulfate particles
f_{ws}	fraction of the particle's weight, which is composed of sulfur
H	scale height of CO ₂
h_c	heating rate due to carbon dioxide
h_s	induced cooling rate due to sulfur dioxide
k	empirical constant relating N_d and N_c
m_a	mass of aerosol [grams/cm ³]
m_c	mass of aerosol particles that participate in CCN formation [grams/cm ³]
m_{co}	mass of carbon dioxide [grams/cm ³]
m_s	mass of sulfur [grams/cm ³]
M_c	atomic weight of carbon
M_{co}	molecular weight of carbon dioxide
M_s	atomic weight of sulfur
n_c	number of carbon atoms in a unit mass of coal
n_s	number of sulfur atoms in a unit of mass of coal
N_a	aerosol concentration [aerosols/cm ³]
N_c	cloud condensation nuclei concentration [CCN's/cm ³]
N_d	cloud droplet concentration [drops/cm ³]
N_s	concentration of sulfur atoms or SO ₂ molecules [atoms or molecules/cm ³]
r_g	particle number distribution median radius [μm]
r_m	particle mass distribution mean radius [μm]
r_{min}	minimum particle radius that can form a cloud drop for supersaturation S
R_c	increment in the CO ₂ emission rate [molecules per cm ² day ⁻¹]
R_s	increment in the SO ₂ emission rate [molecules per cm ² day ⁻¹]
S	supersaturation in percent
T_a	lifetime of an aerosol particle in the atmosphere [days]
T_c	effective lifetime of CO ₂ [years]
V_a	particle volume for radius r_m [μm]
V_m	volume of one mole (22 400 cm ³)
α	fraction of the earth's surface covered by industrial pollution
β	fraction of the earth's surface covered by low clouds, which can be effected by pollution
γ	coefficient relating change in cloud albedo with a relative change in optical thickness
η	ratio of indirect cooling by SO ₂ molecules to heating by CO ₂ molecules
ξ	ratio of cooling to heating due to the burning of coal
τ_c	cloud optical thickness

6 and 7 shows that for biomass burning, heating is more probable than cooling.

5. Discussion and conclusions

The predictions of the effect of an increase in coal or oil burning, presented in Table 6, show that within the present conditions the cooling effect from coal and oil burning may range from 0.4 to 8 times the heating

effect. Within this large uncertainty, we conclude that it is presently more likely that fossil fuel burning causes cooling of the atmosphere than heating it. Biomass burning associated with deforestation, on the other hand, has a much higher probability of heating the atmosphere than cooling it. Future increases in coal and oil burning may saturate the cooling effect and allow the heating to dominate. For example, a doubling of CO₂ may generate heating that is 2–16 times larger than cooling. Therefore, if SO₂ indirect cooling has not thus far caused a cooling trend of the earth's surface (as is indicated for a wide range of possible scenarios in Table 6) then it may delay the heating effect, "waiting" temporarily for the saturation of the cooling effect. An interesting paradox is to consider the situation that civilization may find itself in, if in the future an alternative, nonpolluting energy source is found. A sudden drop in coal and oil consumption will eliminate the cooling effect in a matter of days (aerosol lifetime is around 5 days), while the CO₂ heating will continue for additional decades, thus increasing the global temperature more drastically than before the drop in the fossil fuel use. Note that recent efforts to suppress SO₂ emission in order to prevent acid rain may increase the net heating due to fossil fuel consumption.

Several effects were not accounted for in this analysis. For example, the cooling effect may be larger if clouds of smaller droplets will also result in less precipitation, and therefore in longer lifetimes (in addition to their higher albedo—see Hobbs and Rangno 1985; Albrecht 1989; Radke et al. 1989). Since most SO₂ is generated and remains over land, it should affect the radiative balance over land much more than over oceans. The smaller thermal inertia of land and the limited land-ocean thermal coupling (Thompson and Schneider 1989) will cause a stronger effect on the higher land temperature than on the ocean temperature and will result in smaller thermal emission to space. The reduction in thermal emission will tend to reduce the cooling effect (Schneider, private communication). Detailed account of this effect requires the use of climate models (Kaufman and Chou, in preparation). In addition, the persistence of maritime stratiform clouds over the eastern portions of the oceans may also limit the cooling effect. As a result of the prevailing westerly winds, these cloud layers are not affected substantially by aerosol pollution (Ramanathan et al. 1985; Charlson et al. 1987).

It is possible that the present discussion will further confuse any public strategy concerning the use of fossil fuel and biomass burning. The main conclusion from this analysis should be that, although we should acknowledge that the effect of large scale emission of particles and gases into the atmosphere may result in climatic changes, in addition to their effect on atmospheric chemistry (Crutzen 1988), it may be too early to predict the direction and possible twists of the effect. As a result the concept (Schneider 1989) that society

should pursue those actions which reduce fossil fuel use while providing other widely agreed societal benefits is recommended. In this case, even if the change in climate does not materialize, the action will have some societal benefits (e.g., encouraging the development of fuel efficient public transportation).

In order to decrease the uncertainty as to the effect of coal and oil burning on climate, there is a need to verify experimentally the relation between the presence of pollution and the corresponding change in cloud characteristics. Since the effect of pollution on clouds is usually much smaller than the variability in the cloud characteristics due to dynamic effects, the relation must be based on statistical studies of cloud characteristics and aerosol density. Satellite imagery can be used to study thousands of clouds simultaneously (Arking and Childs 1985), parallel to studies of the surrounding aerosol (Kaufman et al. 1990; Setzer 1988; Setzer and Pereira 1989). Because of the nonlinearity of the relation between cloud albedo and aerosol density, in addition to detailed studies of cloud microphysics from aircrafts or balloons, and its relation to aerosol concentration, there is a need to study the relation between the aerosol concentration and the average cloud albedo from satellite imagery.

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