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Does Fossil Fuel Combustion Lead To Global Warming?†

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Abstract - Tropospheric sulfate aerosols produced by atmospheric oxidation of SO_2 emitted from fossil fuel combustion scatter solar radiation and enhance the reflectivity of clouds. Both effects decrease the absorption of solar radiation by the earth-atmosphere system. This cooling influence tends to offset the warming influence resulting from increased absorption of terrestrial infrared radiation by increased atmospheric concentrations of CO_2. The sulfate forcing is estimated to be offsetting 70% of the forcing by CO_2 derived from fossil fuel combustion, although the uncertainty of this estimate is quite large--range 28 to 140%, the latter figure indicating that the present combined forcing is net cooling. Because of the vastly different atmospheric residence times of sulfate aerosol (about a week) and CO_2 (about 100 years), the cooling influence of sulfate aerosol is exerted immediately, whereas most of the warming influence of CO_2 is exerted over more than 100 years. Consequently the total forcing integrated over the entire time the materials reside in the atmosphere is net warming, with the total CO_2 forcing estimated to exceed the sulfate forcing by a factor of 4 (uncertainty range 2 to more than 10). The present situation in which the forcing by sulfate is comparable to that by CO_2 is shown to be a consequence of the steeply increasing rates of emissions over the industrial era.

1. Introduction

It is generally accepted that fossil-fuel combustion is responsible for much of the increase in atmospheric carbon dioxide concentrations above preindustrial values and that this increased CO_2 results in an increased longwave (infrared) radiative forcing of the troposphere that leads to a warming of the earth's surface.[1] Recently attention has been called to a shortwave radiative forcing by tropospheric sulfate aerosols produced by atmospheric oxidation of SO_2 emitted from fossil-fuel combustion.[2-9] These aerosols scatter solar radiation, increasing clear-sky planetary reflectivity. Increased concentrations of aerosol particles also lead to increased reflectivity of clouds. Both effects are thought to contribute a cooling influence on the planetary energy balance that to some extent offsets the

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warming influence due to increased CO_2. Accurate knowledge of the net forcing due to fossil-fuel combustion is necessary, both for interpretation of temperature trends over the industrial era and for formulation of future policies regarding fossil-fuel combustion.[1, 4, 10-12] It is therefore necessary to examine the climate influence of fossil-fuel combustion, taking into account both the infrared forcing due to increased concentrations of CO_2 and the shortwave forcing due to sulfate aerosol. A major difference in the two forcings is the residence time of the materials in the atmosphere: decades to centuries for CO_2, days to weeks for sulfate. Another key parameter governing the relative magnitudes of these forcings is the sulfur content in the fuel, which has decreased in recent years and which may be expected to decrease further in response to controls on SO_2 emissions aimed at reducing acid deposition.

In this article, I first describe and compare the two modes of radiative forcing and review estimates of the normalized instantaneous radiative forcing due to each of the two substances. I then examine the extent to which the relative magnitudes of warming and cooling depend on the time history of emissions, in view of the great differences in atmospheric residence times of sulfate and CO_2. I show that as a consequence of the greatly differing residence times, the net radiative forcing by the two substances, emitted in constant ratio, can be quite different for different time profiles of emissions. I then apply this analysis to a time history of fossil-fuel combustion over the industrial period to assess the relative influence of these two opposing forcings up to the present time and consider implications for the future.

2. Analysis

2.1. Comparison of present radiative influences of fossil-fuel derived CO_2 and sulfate

It is well established that the mixing ratio of atmospheric CO_2 has been increasing substantially over the industrial era[13] from the preindustrial value of 280 ppm (parts per million by volume) to the present (1990) value of 353 ppm. The radiative influence of this excess CO_2 is quantified as radiative forcing of the earth-troposphere system, which is estimated, again for 1990, as 1.5 $W \cdot m^{-2}$;[14] this is the greatest single contribution to anthropogenic greenhouse gas forcing, which is estimated as 2.45 $W \cdot m^{-2}$ in the aggregate. Although the increase in CO_2 concentration is well established, apportionment of that increase to fossil-fuel combustion and to other activities such as deforestation and land use changes is less certain. The contribution of fossil-fuel combustion to the net increase in CO_2 flux into the atmosphere over the industrial period as a whole is estimated as 63 ± 10% ;[14] this relative contribution has been increasing over this period, with the present (1980-1989) contribution estimated as 76 ± 12%. In view of the long residence time of excess CO_2 in the atmosphere (decades to centuries[14]), it is not clear *a priori* what are the appropriate values to employ in estimating the fossil-fuel contribution to excess CO_2 forcing, so I take this range as 71 ± 17%; the radiative forcing attributed to fossil-fuel CO_2 is thus estimated as 1.07 ± 0.26 $W \cdot m^{-2}$.

The radiative forcing due to anthropogenic sulfate has recently been the subject of much scrutiny. The steady-state global-mean direct radiative forcing was estimated by a box model calculation[9] as -1.3 $W \cdot m^{-2}$, uncertain to a factor of 2, the principal contributions to the uncertainty being the fractional conversion of SO_2 to sulfate and the mean atmospheric residence time of the sulfate aerosol, which is about a week. (The negative sign indicates that this radiative influence is one of cooling, in contrast to the warming influence of excess CO_2). Calculations using sulfate concentration fields obtained with a three dimensional global model[15] led to an estimate of global mean forcing of -0.60 $W \cdot m^{-2}$,[7] again uncertain to a factor of 2. In addition to the direct forcing, Charlson et al[9] estimated the so-called indirect forcing due to enhancement of cloud albedo, based on an assumed 15% global-mean increase in the concentration of droplets in marine stratus clouds, that in turn is based on a comparison of measured sulfate aerosol concentrations at locations remote from industrial emissions in the Northern and Southern Hemispheres.[3] The resulting global-mean cloud forcing was estimated as -1 $W \cdot m^{-2}$, additive to the direct forcing; however, this estimate must be viewed as even more uncertain than that of the direct forcing because of the highly nonuniform spatial distribution of the aerosol, the unknown dependence of cloud droplet concentration on sulfate concentration, and the nonlinear dependence of cloud reflectance on cloud droplet concentration. Recently Wigley and Raper[4] reviewing the available evidence on sulfate forcing estimated the total forcing (direct plus cloud) as -0.75 ± 0.38 $W \cdot m^{-2}$. Since the focus of the present article is restricted to the effects of fossil-fuel combustion, the above estimates should be reduced by 8% to account for non fossil-fuel anthropogenic sulfur emissions, largely from smelting of non-ferrous metals;[16] with this reduction, the Wigley-Raper[4] estimate for the radiative forcing by fossil-fuel sulfate becomes -0.70 ± 0.35 $W \cdot m^{-2}$.

The several estimates of sulfate forcing range widely. They can be refined only by improved descriptions of the distribution and radiative properties of this aerosol. The sulfate aerosol forcing is distributed quite differently in space and time from the greenhouse gas forcing and consequently equivalent climate response is not necessarily expected. Nonetheless it is clear that this forcing is substantial as measured by the yardstick of fossil-fuel CO_2 forcing or total greenhouse gas forcing. If we take the Wigley-Raper[4] estimate as a conservative recent estimate of the sulfate forcing, and combine the range of this estimate with the range of estimates of fossil-fuel CO_2 forcing given above, then we obtain an estimate for the present net radiative forcing due to fossil-fuel combustion of 0.37 ± 0.61 $W \cdot m^{-2}$, i.e. ranging from + 0.98 $W \cdot m^{-2}$ (net warming) to - 0.24 $W \cdot m^{-2}$ (net cooling).

In view of the substantial uncertainty, even in the sign of the net forcing, it appears that a systematic comparison of the forcings of CO_2 and sulfate is warranted. Further, in view of the widely differing residence times of the two substances in the atmosphere, such a comparison must focus on the time dependence of the forcings by these materials subsequent to their introduction into the atmosphere.

2.2. Global warming potentials and integrals

To compare the radiative forcing of sulfate aerosols to that of CO_2, I employ and extend the global warming potential (GWP) approach,[14, 17] which was developed to facilitate comparison of the climate influence of different greenhouse gases. The GWP is a single index that accounts for the climate influence that would result from introduction of a specified incremental amount of an infrared-active gas into the atmosphere, relative to that for carbon dioxide. As generally employed[14] the GWP of a substance i is a dimensionless quantity evaluated as the time integrated radiative forcing of the earth-troposphere system due to introduction of a unit incremental amount of the substance into the atmosphere, ratioed to the same quantity for CO_2:

$$\mathrm{GWP}_i(T) \equiv \int_0^T a_i f_i(t)\,dt \bigg/ \int_0^T a_{\mathrm{CO_2}} f_{\mathrm{CO_2}}(t)\,dt\,. \tag{1}$$

Here a_i represents the instantaneous radiative forcing due to an incremental amount (1 kg) of substance i in the atmosphere, and $f_i(t)$, the decay profile of the substance, is the fraction of material remaining in the atmosphere at a time t subsequent to its introduction. The integral is taken over a period of time T that is denoted the "time horizon" of concern; if the substance has a different decay profile in the atmosphere from that of CO_2, the GWP will depend on the time horizon that is employed. This useful quantity encompasses the effects of both the magnitude of instantaneous radiative forcing and residence time of the substance in describing its climatic influence. Although criticized as not accurately representing differences in the details of forcings by different greenhouse gases,[18] the approach continues to be employed[10] as a useful first-order estimator of total global influences.

It is seen that the GWP of a particular substance i depends not only on the radiative and persistence properties of that substance, but also on the corresponding properties of CO_2. In view of the vastly differing time scales characterizing the removal of CO_2 and aerosol sulfate from the atmosphere, the two integrals comprising the numerator and denominator of the GWP exhibit quite different time behavior. Consequently much more insight can be gained by examining the two integrals individually than can be gained from examination only of the ratio. Moreover uncertainties in the properties of both the substance and CO_2 contribute to uncertainties in the GWP. As it turns out there are substantial uncertainties in the atmospheric decay profile not only of sulfate aerosol but also of excess CO_2. Indeed, the Intergovernmental Panel on Climate Change (IPCC) states that because of uncertainties associated with specifying the lifetime of CO_2, "presentation of the GWP relative to CO_2 may not be the ideal choice."[14] For these reasons I introduce a quantity that I denote as the Greenhouse Warming Integral (GWI)

$$\mathrm{GWI}_i(T) \equiv \int_0^T a_i f_i(t)\,dt\,. \tag{2}$$

Unlike the GWP, the dependence of the GWI on the time horizon T depends on the residence time profile $f_i(t)$ of only a single substance. In contrast to the GWP, the GWI has dimension; for a_i expressed in units $\mathrm{W{\cdot}m^{-2}{\cdot}kg^{-1}}$, and for time taken in seconds, the GWI has units $\mathrm{J{\cdot}m^{-2}{\cdot}kg^{-1}}$. In the limit $T \to \infty$ the GWI admits to a ready interpretation: it is the global-mean incremental heating per unit surface area of the earth resulting from the introduction of 1 kg of the substance i into the atmosphere. If the forcing per kilogram a_i is a constant this total GWI is simply expressed as the product of a_i times the mean residence time of the material in the atmosphere τ_i, i.e.

$$\mathrm{GWI}_i^{\infty} = a_i \int_0^{\infty} f_i(t)dt = a_i \tau_i , \tag{3}$$

where τ_i is the mean residence time of the material in the atmosphere. For a gas whose decay profile is exponential, the partial GWI for time horizon T is $a_i \tau_i [1 - f_i(T)] = a_i \tau_i [1 - \exp(-T/\tau_i)]$.

For a species whose residence time is sufficiently short relative to secular changes in source strength that it may be considered to be in steady state, the total GWI may equivalently be calculated as the global-mean incremental radiative forcing F_i (W·m^{-2}) divided by the total global source strength Q_i (kg s^{-1}) of the substance,[19] viz.

$$\mathrm{GWI}_i^{\infty} = F_i / Q_i \tag{4}$$

The incremental forcing per kilogram of material emitted into the atmosphere can thus be evaluated from Eq. (3) as

$$a_i = F_i / Q_i \tau_i \tag{5}$$

The GWP approach has previously been applied only to greenhouse gases, and in its 1992 update the IPCC explicitly indicated that no attempt was made to define a GWP for anthropogenic sulfate emissions, in view of "the very different character of the forcing due to aerosol sulphate as compared to that of the greenhouse gases."[20] In fact, however, the approach is readily extended to aerosol sulfate by means of a sign convention such that a negative GWP or GWI denotes a cooling influence. The extension appears to be useful, and I make use of it here in order to compare the magnitude of radiative forcing of climate by fossil-fuel CO_2 and sulfate aerosol.

2.3. Global warming integral for sulfate aerosol

To derive a GWI for sulfate I use the radiative forcing estimates noted above together with the corresponding emissions of anthropogenic SO_2. Scattering of shortwave radiation (direct forcing) is to good approximation linear in emitted SO_2 (the extent of SO_2 oxidation is assumed linear in SO_2 concentration, the resulting sulfate aerosol is optically thin, and the removal of the aerosol from the atmosphere, mainly by precipitation, is assumed independent of the aerosol concentration). The Charlson et al[9] box model calculation yielded, for annual anthropogenic emissions of SO_2, Q_S, taken as 90 Tg S yr^{-1} and sulfate mean residence time of 7 days, a steady-state global-mean direct radiative forcing of -1.3 W·m^{-2}. The three-dimensional global model of Langner and Rodhe[15] led to an estimate[7] of the steady-state global-mean direct radiative forcing of -0.60 W·m^{-2} for the SO_2 emission rate taken as 71 Tg S yr^{-1}; in these calculations the mean residence time of sulfate aerosol was found to be about 5 days. From these estimates and Eqs. (4) and (5) it is possible to evaluate the total (infinite time) GWI of tropospheric sulfate aerosol, and the instantaneous average forcing per kilogram emitted sulfur, a_S, as given in Table 1.

Table 1. Comparison of GWIs and related quantities describing shortwave radiative forcing by sulfate aerosol evaluated with forcing calculated with a box model and with a three-dimensional global model. Entries in parentheses in the fourth column, not employed in the present evaluations, are given to permit comparison.

	Direct Forcing			Cloud Forcing
Quantity	Box Model (Ref. 9)	3-D Global Model (Ref. 7)	Employed in present evaluations	Box Model (Ref. 9)
Q_S, Tg S yr^{-1}	90	71	(81)	90
Q_S, kg S s^{-1}	2.85×10^3	2.25×10^3	(2.58×10^3)	2.85×10^3
F_S, W·m^{-2}	-1.3	-0.6	(-1.0)	-1
GWI$_S^\infty$, μJ·m^{-2}·kg^{-1}	-460	-270	-360	-360
$\tau_{SO_4^{2-}}$, days	7	5	6	7
$\tau_{SO_4^{2-}}$, s	6.05×10^5	4.32×10^5	5.18×10^5	6.05×10^5
a_S, pW·m^{-2}·kg^{-1}	-750	-620	-690	-580

Several points should be noted regarding this table. First, the GWI is negative, as is a_S, indicative of a cooling influence. Second, the factor of two difference in forcing estimated by the two models is seen to be due in large part to the different emission rates and mean residence times employed; the actual forcing per kilogram of sulfur emitted into the atmosphere, a_S, differs between the models by only about 20%. However, this agreement should not necessarily be taken as indicative of the certainty of the estimates. The estimates of GWI$_S$ and a_S should be viewed as uncertain to roughly a factor of two. For the purpose of the present comparison of radiative forcing of sulfate and CO_2 from fossil-fuel combustion, I employ the values -360 μJ·m^{-2}·kg^{-1} and -690 pW·m^{-2}·kg^{-1}, respectively, corresponding to a mean residence time of sulfate of 6 days. These values, together with the present (1989) rate of emissions of SO_2 from fossil-fuel combustion (87 Tg S yr^{-1}, as inferred from the present rate of CO_2 emissions, 5.8 Pg C yr^{-1}, and the present sulfur to carbon ratio in fossil-fuel, 0.014, Fig. 1) yield for the present global average direct forcing by sulfate aerosol -1.0 W·m^{-2}, essentially the same magnitude as the estimate given above for present radiative forcing by fossil-fuel derived CO_2. As improved estimates of the several quantities become available, the GWI estimates obtained here may be scaled appropriately.

In addition to the direct forcing there is also thought to be a contribution to the GWI from the cloud forcing. Charlson et al[9] estimate this forcing as -1 W·m^{-2} but highly uncertain. For reference, the corresponding contributions to GWI and a_S are also given in Table 1. However, in view of the uncertainty in estimating the cloud forcing, I use only the direct forcing due to clear-sky light scattering in comparisons with the CO_2 forcing; the resulting comparisons can be scaled appropriately to include the cloud forcing.

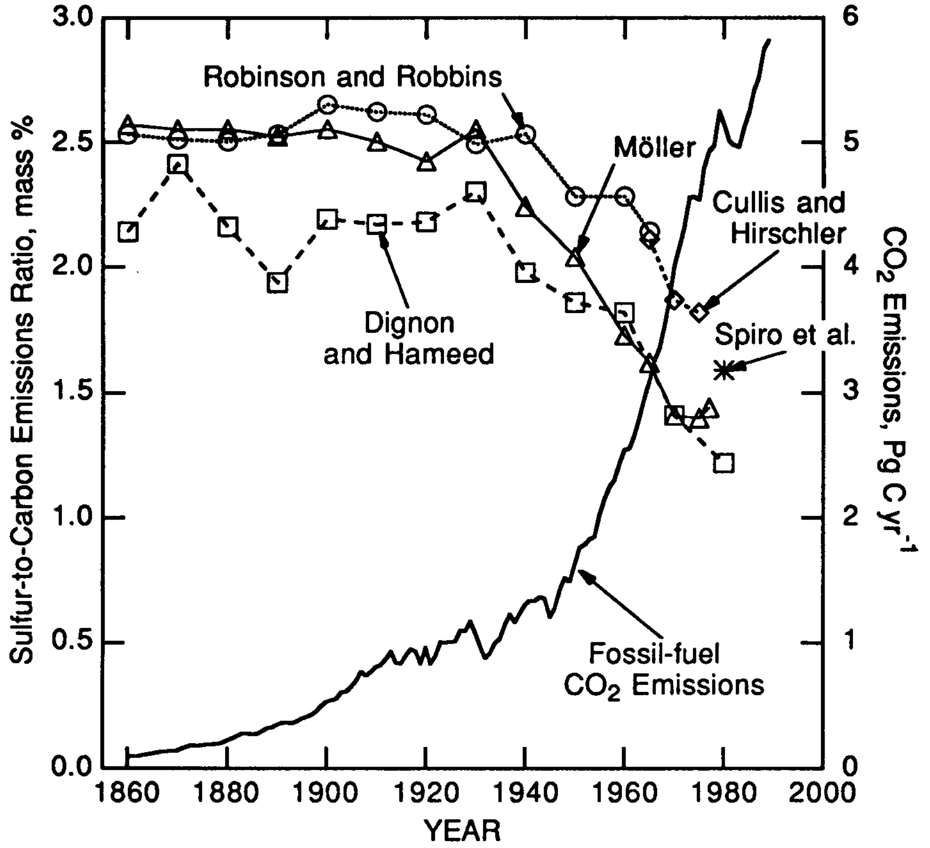

Fig. 1. Time history of the ratio of sulfur-to-carbon emissions from fossil-fuel combustion evaluated from Refs. 16, 24-27 (left axis) and the rate of CO_2 emissions from fossil-fuel combustion[28] (right axis).

2.4. Global warming integral for CO_2

The incremental longwave radiative forcing of the troposphere by CO_2 is approximately logarithmic in CO_2 concentration, such that a doubling of CO_2 concentration corresponds to an incremental forcing of approximately 4 $\mathrm{W{\cdot}m^{-2}}$. Thus the instantaneous forcing of an incremental amount of CO_2 added to the atmosphere $\Delta[CO_2]$ can be approximated as[14]

$$a_{CO_2} = \left(\frac{4\ \mathrm{W \cdot m^{-2}}}{\ln 2}\right)\Delta \ln[CO_2] = \left(5.8\ \mathrm{W \cdot m^{-2}}\right)\frac{\Delta[CO_2]}{[CO_2]}, \qquad (6)$$

where $[CO_2]$ is the amount initially present in the atmosphere. For $[CO_2]$ taken as 670 Pg C, equivalent to 315 ppm, intermediate between preindustrial and present values, $a_{CO_2} = 8.6 \times 10^{-15}\ \mathrm{W{\cdot}m^{-2}}$ per kg C. Because the departure from linearity over this range of $[CO_2]$ is fairly small, the GWI of CO_2 is adequately approximated as $a_{CO_2}\int_0^T f_{CO_2}(t)dt$.

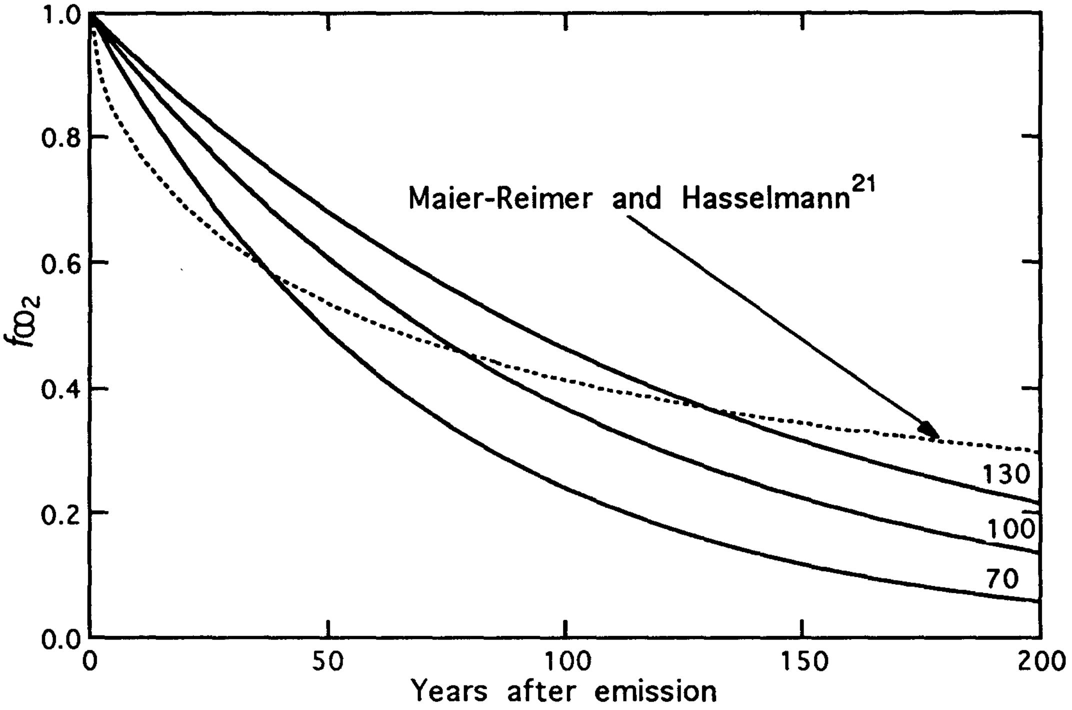

Fig. 2. Profiles of the fraction of excess CO_2 remaining in the atmosphere as functions of time subsequent to introduction into atmosphere. The solid curves show exponential decays for the indicated mean residence times in years. The dotted non-exponential decay curve was obtained with the multi-component box model employed in Global Warming Potential estimates by IPCC.[14]

In contrast to sulfate, excess CO_2 from fossil-fuel combustion remains in the atmosphere for decades to centuries. The decay profile of excess CO_2 in the atmosphere is not well established. Multi-compartment model calculations[21,22] indicate a nonexponential decay, with an initial half-life of about 50 yr but with the second half-life much greater, about 250 yr, Fig. 2; in fact, according to such models, the incremental CO_2 mixing ratio may never return to zero, depending on assumptions of the ultimate geochemical fate of incremental CO_2 added to the atmosphere. Such a profile, for which the equivalent mean residence time was stated to be approximately 120 years, was employed by the IPCC[1,10] in GWP estimates.

Alternative estimates of the mean residence time of CO_2 in the atmosphere can be inferred from the time dependence of CO_2 concentration in the atmosphere in excess of the preindustrial value, the rate of increase in atmospheric CO_2, and the rate of CO_2 emissions from industrial activity as a function of time. Using the data given by IPCC for these quantities, I infer the mean residence time of excess CO_2 in the present atmosphere to be about 85 years. A value of 68 years can be inferred from estimates of carbon fluxes to surface ocean water derived from the rate of decrease of ^{13}C concentration.[23] In view of this range of values, I take the mean residence time as 100 years, uncertain to 30%; the corresponding exponential decay profiles are given in Fig. 2. It is seen that these profiles decay initially more slowly than for the nonlinear model, but that ultimately they decay much more rapidly. The total GWI (time horizon $T \rightarrow \infty$) is quite sensitive to the long-time behavior of these profiles.

We are now prepared to evaluate GWI(T) for CO_2. Fig. 3 shows this quantity for the three exponential decay profiles (τ_{CO_2} = 70, 100, 130 years) and for the nonexponential profile of Maier-Reimer and Hasselmann.[21] As expected, these profiles increase from zero over a time scale of some 200 years. The final value for the exponential profiles is GWI_C^{∞} = (27 ± 8) $\mu J \cdot m^{-2}$ per kg C emitted into the atmosphere as CO_2; for the nonexponential decay profile the value is greater and in fact may continue to increase without bound, depending on assumptions regarding the ultimate fate of incremental CO_2.

Because the time scale of the decay of excess CO_2 in the atmosphere is long compared to that characterizing the increase in the rate of emission of CO_2 over the industrial period, (Fig. 1), atmospheric CO_2 cannot be considered to be in steady state; indeed the annual increase in the burden of atmospheric CO_2 is a substantial fraction of the annual fossil-fuel

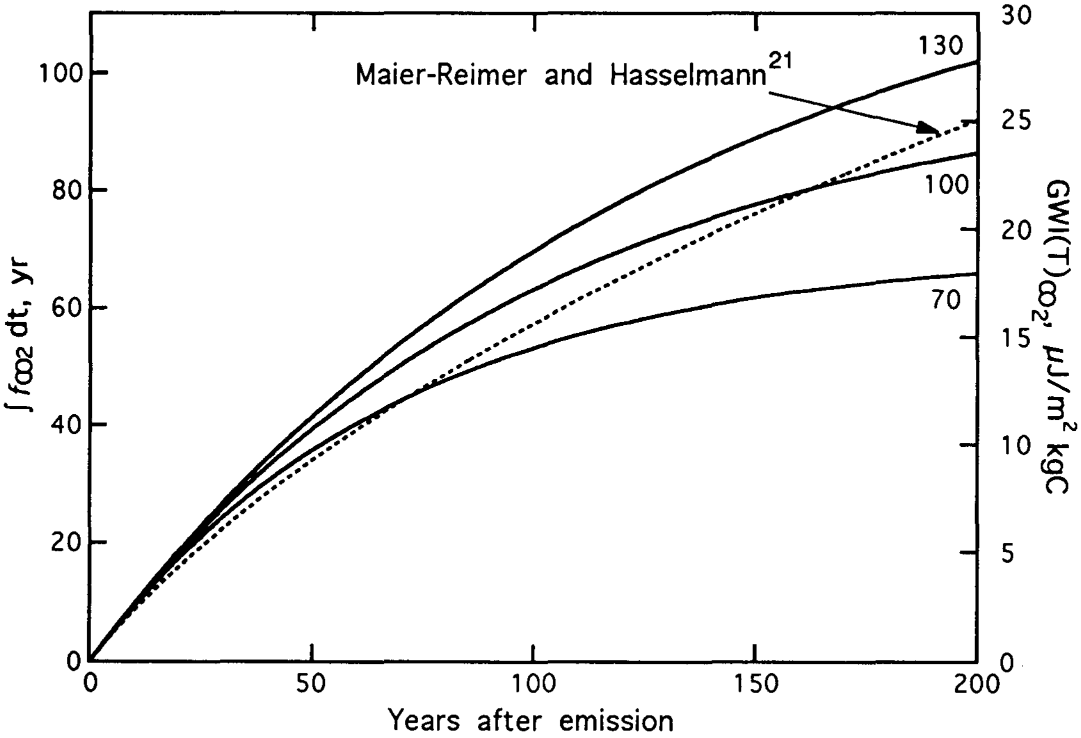

Fig. 3. Profiles of the time integrals of estimates of the fraction of excess CO_2 remaining in the atmosphere as a function of time subsequent to introduction into atmosphere. The solid curves show the integrals of exponential decay for the indicated mean residence times in years. The dotted curve shows the integral of the non-exponential decay curve obtained with the multi-component box model. The value of the curves indicated on the right-hand axis is the Global Warming Integral evaluated for instantaneous radiative forcing due to an incremental amount of CO_2 in the atmosphere, a_{CO_2}, equal to 8.6×10^{-15} $W \cdot m^{-2}$ per kg C.

emissions. For this reason, it is mandatory to consider the time dependence of emission and forcing in comparing the climate forcing of sulfate aerosol and CO_2. In the long run the relative contributions of the two substances can be compared by consideration of the infinite-time integrals. However, on a time scale over which CO_2 is not in steady state, it is necessary to employ the partial GWI for CO_2 and to take explicit account of the historical time profile of CO_2 emissions. As shown below, this is crucial for comparison of the two forcings over the industrial period.

2.5. Comparison of infinite time GWIs of CO_2 and sulfate aerosol

With the total GWIs for CO_2 and sulfate it is possible to compare the net radiative forcing of the two species, that is the net GWI for fossil-fuel combustion, integrated over the entire residence times of the two materials in the atmosphere. Here I choose to express this forcing in terms of the unit mass emissions of carbon, expressing the emission of sulfur in terms of the mass ratio in emissions (Q_S/Q_C). Thus, the net GWI for fossil-fuel combustion normalized to emission of carbon is

$$\mathrm{GWI}_{\mathrm{ff}}^{\infty} = \mathrm{GWI}_{\mathrm{C}}^{\infty} + (Q_S / Q_C)\mathrm{GWI}_{\mathrm{S}}^{\infty}. \tag{7a}$$

For the two GWIs evaluated by using Eq. (3), we have

$$\mathrm{GWI}_{\mathrm{ff}}^{\infty} = a_{CO_2}\tau_{CO_2} + (Q_S / Q_C)a_S\tau_{SO_4^{2-}}. \tag{7b}$$

Equation (7) permits determination of a critical ratio of emissions of SO_2 and CO_2, $(Q_S/Q_C)^*$, such that the incremental forcings negate each other, i.e. such that $\mathrm{GWI}_{\mathrm{ff}}^{\infty} = 0$ as

$$(Q_S / Q_C)^* = -\frac{\mathrm{GWI}_{\mathrm{C}}^{\infty}}{\mathrm{GWI}_{\mathrm{S}}^{\infty}}, \tag{8}$$

or by Eq. (7b)

$$(Q_S / Q_C)^* = -\frac{a_{CO_2}\tau_{CO_2}}{a_S\,\tau_{SO_4^{2-}}} \tag{8b}$$

For $(Q_S/Q_C) < (Q_S/Q_C)^*$ the net forcing is positive, i.e. warming whereas, for $(Q_S/Q_C) > (Q_S/Q_C)^*$, the net forcing is negative. For the total GWIs obtained above $(Q_S/Q_C)^*$, is equal to 0.075, with an uncertainty, due largely to the uncertainty in the GWI of sulfate, of about a factor of 2. This value is roughly four times the mass ratio of sulfur to carbon in emissions from fossil-fuel combustion (Fig. 1); the emission-weighted average over the period 1860 to 1987, $\overline{(Q_S / Q_C)}$, is 0.019. Consequently, when the CO_2 forcing over its entire residence time in the atmosphere is considered, the positive forcing due to CO_2 exceeds the negative forcing due to sulfate aerosol. It follows that the net effect of fossil-fuel combustion, evaluated for the total period of time over which the excess CO_2 remains in the atmosphere, is unequivocally one of heating. This result would also apply to the steady-state emissions scenario, that is, to a situation in which both substances are emitted into the atmosphere at constant rate for a period long compared to the mean residence time of excess CO_2. It is clear, however, that the conclusion might not be the same with respect to the actual net forcing experienced to date over the industrial era, especially as the rate of emissions has been increasing strongly over this period, and therefore much of the ultimate heating influence of excess CO_2 still present in the atmosphere has not yet been exerted.

2.6. Time-dependent global warming integral for fossil fuel

It should be apparent by now that the net radiative forcing experienced by the earth-troposphere system due to CO_2 and sulfate aerosol resulting from fossil fuel combustion must be considered as a function of time following emission. Insight into this time dependence may be gained by considering the time-dependent GWI for fossil fuel containing a specified mass ratio of sulfur to carbon. By analogy to Eq. (7) this time-dependent GWI, normalized to 1 kg C, is given as

$$\mathrm{GWI}_{\mathrm{ff}}(T) = \mathrm{GWI}_{\mathrm{C}}(T) + (Q_{\mathrm{S}}/Q_{\mathrm{C}})\mathrm{GWI}_{\mathrm{S}}^{\infty}, \tag{9}$$

where the infinite-time GWI is employed for sulfate because of the short time required to reach that limit. Fig. 4 shows $\mathrm{GWI}_{\mathrm{ff}}(T)$ for $Q_S/Q_C = 0.019$, evaluated for the four GWIs for CO_2 presented in Fig. 3 and for $\mathrm{GWI}_{\mathrm{S}}^{\infty}$ taken as -360 $\mu J \cdot m^{-2} \cdot kg^{-1}$; also shown are GWIs evaluated for values of $\mathrm{GWI}_{\mathrm{S}}^{\infty}$ taken as half and twice that value. It is seen that the GWIs are

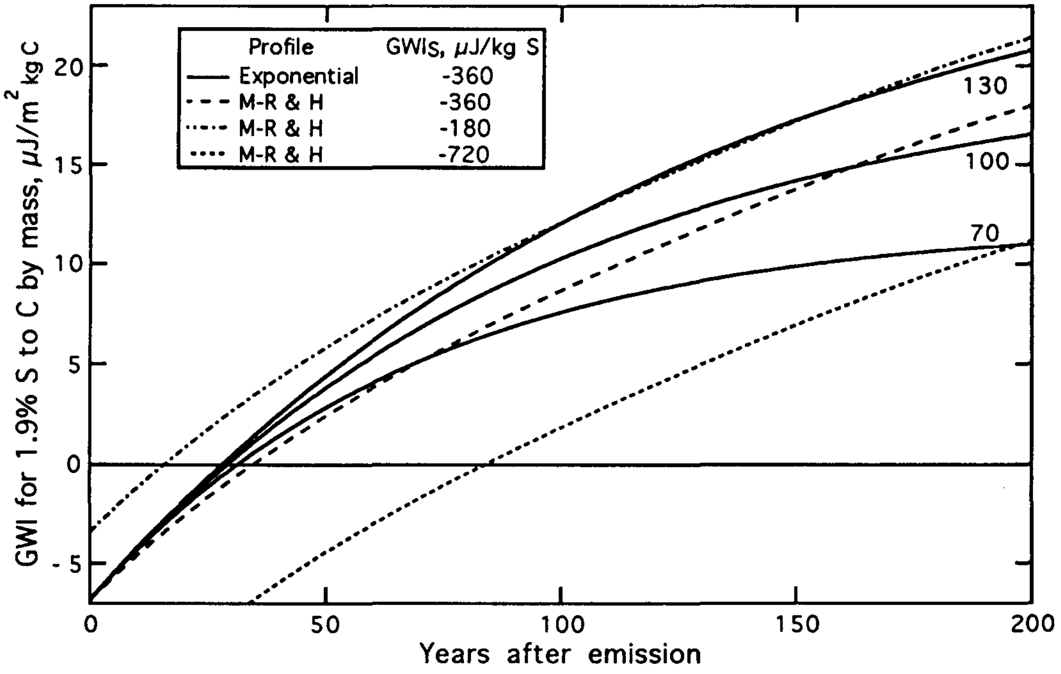

Fig. 4. Profiles of the time-dependent GWI for fossil fuel containing 1.9% S to C by mass, evaluated for several values of $\mathrm{GWI}_{\mathrm{S}}^{\infty}$, and for several decay profiles of CO_2.

simply displaced downward from the corresponding values in Fig. 3, by an amount corresponding to the second term in Eq. (9). Since the GWI for CO_2 is initially quite small and reaches its maximum value only over decades, this displacement has the effect of making GWI_{ff} initially negative; that is the net radiative influence of fossil fuel combustion is intially cooling. The time required for GWI_{ff} to become positive, i.e. net warming, depends only weakly on the choice of the CO_2 profile employed. For GWI_S^∞ taken as -360 $\mu J \cdot m^{-2} \cdot kg^{-1}$, this time is about 30 years. However, the time required for the net influence to become positive is quite sensitive to the value of GWI_S^∞, ranging from 15 years for $GWI_S^\infty = -180$ $\mu J \cdot m^{-2} \cdot kg^{-1}$ to 84 years for $GWI_S^\infty = -720$ $\mu J \cdot m^{-2} \cdot kg^{-1}$. This figure demonstrates the necessity of considering time-dependent GWIs in examining the climate influence of fossil fuel combustion over the industrial period, especially under the circumstance where the rate of fossil fuel combustion has been increasing rapidly on a time scale comparable to the period over which the net GWI changes sign.

2.7. Model for comparing CO_2 and sulfate forcing with exponentially increasing emissions

Here I explore the implications of a CO_2 emission profile that is increasing rapidly relative to the time scale of the decay profile of excess CO_2 in the atmosphere. By considering an exponential growth profile for emissions and an exponential decay profile for atmospheric CO_2, one obtains a great simplification in the algebra that leads to insight. We have already seen that the decay profile of CO_2, f_{CO_2}, appears to be fairly accurately represented by an exponential, with *e*-folding time of about 100 years. Fig. 5 shows that the time history of emissions of fossil-fuel CO_2 over the industrial period is fairly accurately represented by exponential growth with an *e*-folding time of 30 years.

I now evaluate the radiative forcing of a species *i* that is emitted at an exponentially increasing rate $Q_i(t') = Q_i(t_0)\exp[(t'-t_0)/\tau_e]$ and exhibits an exponential decay profile $f_i(t) = \exp[-(t-t')/\tau_i]$. The radiative forcing at time *t* is given by

$$\begin{aligned} F_i(t) &= a_i \int_{-\infty}^{t} Q_i(t') f_i(t-t') dt' \\ &= a_i Q_i(t) \left(\frac{\tau_i \tau_e}{\tau_i + \tau_e} \right). \end{aligned} \tag{10}$$

It is seen that the forcing exerted at any given time t is in constant proportion to the emission rate at that time and thus, by analogy to Eq. (4), I define an effective GWI for this situation as

$$\mathrm{GWI}_i^{\mathrm{exp}} \equiv F_i(t)/Q_i(t) = a_i \left(\frac{\tau_i \tau_e}{\tau_i + \tau_e} \right). \tag{11}$$

This expression differs from that for GWI_i^∞ in that the mean atmospheric residence time τ_i is replaced by the quantity in brackets, the harmonic sum of τ_i and the *e*-folding time of emissions τ_e. For sulfate aerosol, for which $\tau_{SO_4^{2-}} << \tau_e$, this quantity equals $\tau_{SO_4^{2-}}$ and

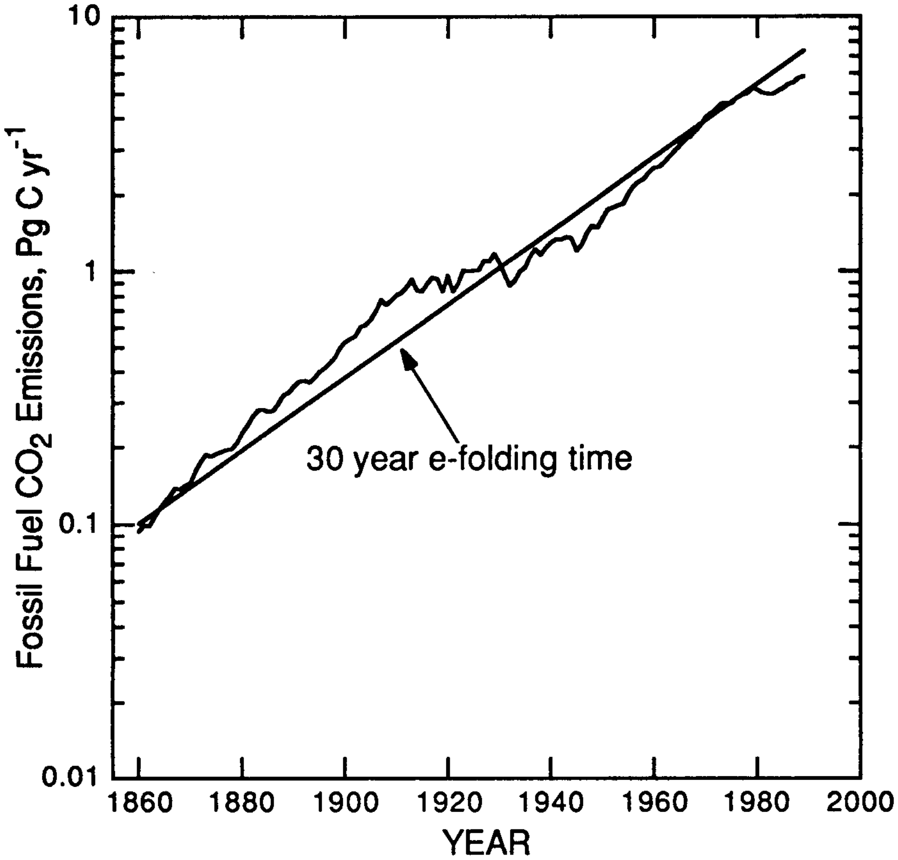

Fig. 5. Rate of anthropogenic CO_2 emissions from fossil-fuel combustion over the industrial era.[28] The straight line represents exponential growth with a 30-year *e*-folding time.

$$\mathrm{GWI}_{\mathrm{S}}^{\mathrm{exp}} = a_{\mathrm{S}}\tau_{\mathrm{SO}_4^{2-}}, \tag{12a}$$

that is, $\mathrm{GWI}_{\mathrm{S}}^{\mathrm{exp}}$ is identical to $\mathrm{GWI}_{\mathrm{S}}^{\infty}$. For CO_2, the situation is nearly reversed, i.e. the atmospheric residence time of excess CO_2, τ_{CO_2}, is almost sufficiently greater than τ_e that the latter can be neglected in the summation in the denominator of Eq. (10). If that were the case, we would have

$$\mathrm{GWI}_{\mathrm{C}}^{\mathrm{exp}} = a_{\mathrm{CO}_2}\tau_{\mathrm{e}}. \tag{12b}$$

In fact, this is not strictly accurate and Eq. (11b) must be replaced by

$$\mathrm{GWI}_{\mathrm{C}}^{\mathrm{exp}} = a_{\mathrm{CO}_2}\tau_{\mathrm{e}}(1+\tau_{\mathrm{e}}/\tau_i)^{-1}, \tag{12c}$$

where for $\tau_e = 30$ years and $\tau_{\mathrm{CO}_2} = 100$ years the correction factor is about 0.77, yielding a value of about 23 years. It is very insightful to compare Eq. (12) with Eq. (3). In a situation of exponentially increasing emissions, with *e*-folding time associated with the increase of emissions τ_e, the mean atmospheric residence time τ_{CO_2} for the long-lived CO_2 has effectively been replaced by the considerably smaller τ_e [in fact, by the even slightly smaller quantity $\tau_e(1+\tau_e/\tau_{CO_2})^{-1}$]. For τ_{CO_2} = 100 ± 30 years, this has the effect of reducing GWI_C from 27 ± 8 to 6.2 ± 0.5 $\mu J \cdot m^{-2}$ per kg C. As may be seen from Fig. 3, this value corresponds to the GWI(T) for a value of T of about 23 years.

In contrast to the situation for CO_2, for the short-lived sulfate there is no difference between the GWC evaluated for the exponentially increasing emissions and the total GWC. This fact implies that for a given emission ratio (Q_S/Q_C), the magnitude of the warming effect of CO_2 relative to the cooling effect of sulfate is substantially reduced from that which would be experienced over the entire residence times of the materials, as evaluated above. The net time-dependent forcing due to fossil-fuel combustion can be evaluated as

$$\begin{aligned} F_{ff}(t) &= F_C(t) + F_S(t) \\ &= Q_C(t)\left[\mathrm{GWI}_C^{exp} + (Q_S/Q_C)\mathrm{GWI}_S^{exp}\right] \\ &= Q_C(t)\left[a_{CO_2}\tau_e(1+\tau_e/\tau_{CO_2})^{-1} + (Q_S/Q_C)a_S\tau_{SO_4^{2-}}\right] \\ &\approx Q_C(t)\left[a_{CO_2}\tau_e + (Q_S/Q_C)a_S\tau_{SO_4^{2-}}\right]. \end{aligned} \tag{13}$$

where as above Q_S is expressed in terms of the mass ratio in emissions (Q_S/Q_C). Also as above we evaluate a critical emissions ratio such that the net forcing is zero [cf. Eq. (8)], i.e.:

$$(Q_S/Q_C)^* = -\frac{\mathrm{GWI}_C^{exp}}{\mathrm{GWI}_S^{exp}} = -\frac{a_{CO_2}\tau_e(1+\tau_e/\tau_{CO_2})^{-1}}{a_S\ \tau_{SO_4^{2-}}} \approx -\frac{a_{CO_2}\tau_e}{a_S\ \tau_{SO_4^{2-}}}. \tag{14}$$

As a consequence of the change in the effective GWI for CO_2, the value for $(Q_S/Q_C)^*$ of 0.075 obtained above with the total GWIs is decreased, for τ_{CO_2} = 100 years, by a factor of 0.23, i.e. to 0.017, again with an uncertainty estimate of a factor of 2. This value is essentially the same as the emission-weighted average $\overline{(Q_S/Q_C)}$ over the period 1860 to 1987, 0.019. To the extent that this model is applicable, this calculation suggests that the net radiative forcing by fossil-fuel combustion emissions, at any time during the exponential growth period, is near zero.

This analysis shows that for an exponentially increasing emissions profile, similar to that actually experienced over the industrial era, the net global warming influence of the two species, having two quite different atmospheric residence times, is quite different from that which would be expected on the basis of the infinite-time global warming integrals. The other side of the coin, however, is the implication that for the values τ_{CO_2} = 100 years and τ_e = 30 years employed in the calculation, only 23% of the total GWI of all the excess CO_2 introduced into the atmosphere according to this emissions profile prior to a given time has been exerted up until that time; the remainder of the GWI of the CO_2 is yet to be exerted, whereas the total GWI for the sulfate is exerted essentially immediately. The fact that the warming influence of CO_2 is to a great extent offset by the cooling influence of sulfate, despite the much greater warming influence of CO_2 than cooling influence of sulfate inferred from the total GWIs, is a subtle consequence of the exponentially increasing emissions profile, i.e. the warming influence of past CO_2 emissions is offset largely by current SO_2 emissions. Such a situation can be maintained indefinitely, but only provided that the emissions of both substances continue to increase at the same exponential rate. However such a pattern of emissions implies that the burden of unexpressed CO_2 forcing also is continuing to increase exponentially. Of course exponential growth rates are hard to sustain indefinitely.

2.8. Comparison of sulfate and CO_2 forcing based on emissions over the industrial era

I now extend the analysis given above to consideration of the forcing resulting from the actual profile of CO_2 and SO_2 emissions from fossil-fuel combustion over the industrial era. This analysis permits comparison of the carbon and sulfur forcings that have occurred only over this period, i.e. excluding the subsequent influence of CO_2 emitted during this period and still remaining in the atmosphere.

From Eq. (3) it may be seen that the integrated forcing exerted over the time period extending from the time of emission t_e to a subsequent time T is given by

$$\mathrm{IF}(T,t_e) = Q_i(t_e)\int_{t_e}^{T} a_i f_i(t)dt = Q_i(t_e)\mathrm{GWI}_i(T,t_e) \qquad (15)$$

Consequently the total forcing exerted over a time period t_0 to T, representing the time period from the beginning of industrialization to the present, is given by

$$\mathrm{IF}(T) \equiv \int_{t_0}^{T} \mathrm{IF}(T,t_e)dt_e = \int_{t_0}^{T} Q_i(t_e)\mathrm{GWI}_i(T,t_e)dt_e\,, \qquad (16)$$

that is, by the integral over time of the GWI weighted by the emission rate. In the case of sulfate, the GWI reaches its final and maximum value $\mathrm{GWI}_\mathrm{S}^\infty$ essentially instantaneously, so that the integrated forcing is equal to the product of $\mathrm{GWI}_\mathrm{S}^\infty$ times the total SO_2 emissions over the period:

$$\mathrm{IF}_\mathrm{S}(T) = \mathrm{GWI}_\mathrm{S}^\infty \int_{t_0}^{T} Q_\mathrm{S}(t_e)dt_e\,. \qquad (17)$$

In turn, this can be expressed in terms of the mean emission ratio $\overline{(Q_\mathrm{S}/Q_\mathrm{C})}$ as

$$\mathrm{IF}_\mathrm{S}(T) = \mathrm{GWI}_\mathrm{S}^\infty\, \overline{(Q_\mathrm{S}/Q_\mathrm{C})}\, E_\mathrm{C}^\mathrm{tot}\,, \qquad (18)$$

where $E_\mathrm{C}^\mathrm{tot}$ is the total emission of CO_2 over the time period,

$$E_\mathrm{C}^\mathrm{tot} = \int_{t_0}^{T} Q_\mathrm{C}(t_e)dt_e\,. \qquad (19)$$

In the case of CO_2, the integral in Eq. (16) must be evaluated for the historical emission rate and an assumed decay profile. It can be expressed as the product of the total emission of CO_2 times the emission-weighted mean GWI, $\langle \mathrm{GWI_C} \rangle$:

$$\mathrm{IF_C}(T) = E_\mathrm{C}^\mathrm{tot} \langle \mathrm{GWI_C} \rangle, \tag{20}$$

where

$$\langle \mathrm{GWI_C} \rangle = \int_{t_0}^{T} \mathrm{GWI_C}(T, t_e)\, Q_\mathrm{C}(t_e)\, dt_e \Big/ \int_{t_0}^{T} Q_\mathrm{C}(t_e) dt_e \,. \tag{21}$$

The net integrated forcing due to fossil-fuel combustion over the time period t_0 to T and exerted over this period is

$$\begin{aligned} \mathrm{IF_{ff}}(T) &= \mathrm{IF_C}(T) + \mathrm{IF_S}(T) \\ &= E_\mathrm{C}^\mathrm{tot} \Big(\langle \mathrm{GWI_C} \rangle + \overline{(Q_\mathrm{S}/Q_\mathrm{C})}\, \mathrm{GWI_S^\infty} \Big) \,. \end{aligned} \tag{22}$$

As above, we define a critical emissions ratio such that the net forcing is zero:

$$(Q_\mathrm{S}/Q_\mathrm{C})^* = -\frac{\langle \mathrm{GWI_C} \rangle}{\mathrm{GWI_S^\infty}} \,. \tag{23}$$

To estimate the mean $\langle \mathrm{GWI_C} \rangle$ for fossil-fuel CO_2 emitted into the atmosphere between 1860 and 1989, I applied the exponential decay model (with $\tau_{CO_2} = 100 \pm 30$ yr) to the fossil-fuel CO_2 emissions profile shown in Fig. 1. This analysis yields $\langle \mathrm{GWI_C} \rangle = 6.4 \pm 0.5$ $\mu J \cdot m^{-2}$ per kg C, essentially identical with the value obtained above with the exponential growth model, 6.2 ± 0.5 $\mu J \cdot m^{-2}$ per kg C. This result supports application of the heuristic exponential growth model given above to the interpretation of the relative forcings of CO_2 and sulfate over the industrial era. The value of $\langle \mathrm{GWI_C} \rangle$ is relatively insensitive to the choice of τ_{CO_2}, an uncertainty in τ_{CO_2} of ± 30% translating into an uncertainty in $\langle \mathrm{GWI_C} \rangle$ of ± 5%. The reason for this is that most of the CO_2 has been emitted in the past 50 years, over which time $\mathrm{GWI_C}$ is quite insensitive to τ_{CO_2} (cf. Fig. 3). In addition to using the exponential decay profile for CO_2, I also evaluated $\langle \mathrm{GWI_C} \rangle$ using the nonexponential decay curve of Maier-Reimer and Hasselmann;[21] the resulting value, 5.7 $\mu J \cdot m^{-2}$ per kg C, is nearly the same as the values obtained with the exponential decay curves. This agreement between the models shows that at least over the time period pertinent to this integration, estimation of the integrated CO_2 forcing is insensitive to the choice of model. The several estimates for $\mathrm{GWI_C}$ are summarized in Table 2.

Because the value of $\mathrm{GWI_C}$ obtained by integration over historical emissions is essentially the same as the value obtained with the model for exponentially increasing emissions, the estimate of $(Q_\mathrm{S}/Q_\mathrm{C})^*$ is also the same as that obtained with that model. That is, the net forcing by CO_2 and sulfate over the industrial era are essentially identical, within the substantial uncertainty of the sulfate forcing. The several estimates for $(Q_\mathrm{S}/Q_\mathrm{C})^*$ are summarized in Table 3.

Table 2. Comparison of GWIs for longwave radiative forcing by CO_2 for exponential and non-exponential models for the decay profile, f_{CO_2}, and for different approaches to the evaluation. The GWIs are expressed in $\mu J \cdot m^{-2}$ per kg C.

Decay Profile Employed	GWI_C^∞	GWI_C^{exp} (30-year *e*-folding time of emissions)	$\langle GWI_C \rangle$ (1860-1989)
Exponential			
$\tau_{CO_2} = 70$ years	19.0	5.71	5.94
$\tau_{CO_2} = 100$ years	27.2	6.27	6.52
$\tau_{CO_2} = 130$ years	35.3	6.62	6.88
Maier-Reimer and Hasselmann[21]	-----	-----	5.71

3. Summary And Conclusions

A formalism involving Global Warming Integrals (GWIs), based on the Global Warming Potential concept, has been introduced to evaluate and compare the contributions to global warming and cooling from emissions of CO_2 and SO_2, respectively. The total GWI is appropriate for steady state emissions and for consideration of the total influence of CO_2 over its entire residence time in the atmosphere, but, in view of the rapid increase in the rate of CO_2 emissions on a time scale short compared to the residence time, this quantity substantially overestimates the influence that fossil-fuel CO_2 has exerted on climate up to the present time. The partial influence up to the present time was evaluated by two models, an exponential growth model (*e*-folding time 30 years) and direct integration over historical

Table 3. Estimates of critical values of mass emissions ratio of sulfur to carbon from fossil-fuel combustion $(Q_S/Q_C)^*$ that would result in zero net radiative forcing of climate, according to different models of time-dependent emissions. Also shown is the average value $\overline{(Q_S/Q_C)}$ from fossil-fuel combustion from 1860-1989. If the actual emissions ratio $\overline{(Q_S/Q_C)}$ exceeds $(Q_S/Q_C)^*$, then the net influence is cooling; if $\overline{(Q_S/Q_C)}$ is less than $(Q_S/Q_C)^*$, the net influence is warming. The uncertainty in $(Q_S/Q_C)^*$ is estimated as a factor of 2. The estimate of $(Q_S/Q_C)^*$ does not include any contribution from enhanced cloud albedo, which would decrease its value. Estimates of $(Q_S/Q_C)^*$ are uncertain to a factor of 2, largely due to uncertainty in the estimate of GWI_S^∞.

	Critical Mass Emissions Ratio $(Q_S/Q_C)^*$	
Model for $(Q_S/Q_C)^*$	Expression	Value
Steady state	$-GWI_C^\infty / GWI_S^\infty$	0.075
Exponential growth ($\tau_e = 30$ years)	$-GWI_C^{exp} / GWI_S^\infty$	0.017
Integrated (1860-1989)	$-\langle GWI_C \rangle / GWI_S^\infty$	0.017
Actual Emissions Ratio (1860-1989)	$\overline{(Q_S/Q_C)}$	0.019

emissions, yielding essentially the same results. The positive CO_2 forcing and the negative sulfate forcing were compared by comparing the critical sulfur to carbon mass ratio in emissions that would result in zero net forcing, $(Q_S/Q_C)^*$, with the actual ratio characterizing fossil-fuel emissions over the industrial era $\overline{(Q_S/Q_C)}$. These comparisons indicate that the positive CO_2 forcing and the negative sulfate forcing, by direct light scattering only, are essentially equal, although the uncertainty in the latter is roughly a factor of two. This equality indicates that the net influence of fossil-fuel effluents on radiative forcing of climate over the industrial period is near zero, consistent with the estimates of present net forcing on which the calculations of GWIs were based. On the other hand, the analysis shows that despite this near-zero present net forcing, the net influence over the entire projected residence time of the fossil-fuel CO_2 in the atmosphere is positive by a substantial margin.

As noted, the GWI for sulfate and therefore $(Q_S/Q_C)^*$, was evaluated for the forcing due only to direct light scattering, neglecting the cloud forcing. This cloud forcing has been estimated to be roughly equal globally to the direct forcing but is considered less certain because of the untested assumptions that led to this estimate. Use of a value for GWI_S that includes both effects decreases estimates of $(Q_S/Q_C)^*$ by a further factor of about 2, i.e. to 0.038 for GWI_C^∞ and to 0.0085 for $\langle GWI_C \rangle$. The latter figure is substantially less than the historical average sulfur-to-carbon emission ratio (0.019), indicative, if it is accurate, that the net radiative forcing resulting from fossil-fuel combustion over this period has been one of cooling, not warming. However, this conclusion must remain tentative in view of the uncertainty in estimates of the radiative forcing of sulfate aerosol,[4, 9] and the issue can be resolved only by decreasing this uncertainty.

Three final points should be noted. First, the calculations were carried out for a global-mean forcing, whereas in view of spatial and temporal differences in these forcings, substantial net short-term regional forcings of differing sign may be expected. Second, because of differing feedbacks in different regions, e.g., a greater water vapor feedback over tropical oceans where the sulfate forcing is minimal, the mean global response to these disparate forcings might not be the same as that to a uniform forcing at the mean value. Third, in contrast to the sulfate forcing, the full effect of the CO_2 emissions has yet to be exerted. As noted previously,[5-8] SO_2 emissions must inevitably decrease and subsequently, as the balance of the CO_2 forcing is exerted, this forcing must result in a much greater positive global-mean forcing than has been experienced thus far.

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