Relative Influence of Visible and Infrared Optical Properties of a Stratospheric Aerosol Layer on the Global Climate

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ABSTRACT

A simple radiative energy balance model has been developed to assess the impact of stratospheric aerosols on the global climate through their effect on the equilibrium global mean surface temperature. With the assumptions that the radiation in the atmosphere can be treated as diffuse radiation and that the effect of the gases in the stratosphere can be approximated by equivalent gray absorbers and scatterers, an analytic expression which depends only on the optical properties of the aerosol and the planetary albedo is derived for the fractional change in the upward flux of terrestrial infrared radiation at the base of the stratospheric aerosol layer. The fractional change in the upward flux of infrared radiation is then directly related to changes in the global mean surface temperature by using existing results of climate model and radiative convective model calculations. Mie theory is used to compute the scattering and absorbing properties of the aerosol for a range of visible and infrared indices of refraction. Sample calculations are presented that show the fractional change in the upward flux of infrared radiation at the base of the layer as a function of particle size for a specified mass concentration of stratospheric aerosols. The results indicate that both small particles (radii $\lesssim 0.05$ μm) and large particles (radii $\gtrsim 1.0$ μm) generally have a greater influence on terrestrial infrared radiation than on incident solar radiation; therefore, these particles contribute to warming at the surface. Particles of intermediate sizes affect the incident solar radiation more strongly than they affect the terrestrial radiation and thereby contribute to cooling at the surface. The results also demonstrate the feasibility of estimating the largest possible surface temperature response to a given increase in the mass concentration of stratospheric aerosols. Calculations were also performed to enable comparison of the results from the present model with those obtained by approximating the effect of an increase in stratospheric aerosols by means of an equivalent reduction in the solar constant. It is shown that the effects of the aerosols on terrestrial radiation must be negligible, and the aerosols must be nonabsorbing at solar wavelengths in order for the results of the present model to agree with those obtained by assuming a reduction in the solar constant.

1. Introduction

With the prospect of extensive future air travel in the stratosphere and with the ever-present possibility of volcanic eruptions, there is a need for studies of the mechanisms by which an increase in the concentration of stratospheric aerosols might affect the earth's climate. Aerosols scatter and absorb both solar and terrestrial radiation. Unlike the absorption of solar radiation by tropospheric aerosols, which is likely to contribute to surface warming, the absorption and scattering of solar radiation by stratospheric aerosols causes a reduction in the flux of solar radiation that reaches the troposphere and thus may lead to cooler surface temperatures (Mitchell, 1971). On the other hand, the absorption and scattering of terrestrial infrared radiation by stratospheric aerosols causes an increase in the downward flux of infrared radiation below the stratosphere and a decrease in the net infrared flux at the top of the atmosphere. If the earth is to maintain its global radiative energy balance, the aerosol-induced changes to the terrestrial radiative fluxes should contribute to a warming of the lower atmosphere.

In much of the work concerning tropospheric aerosols (Mitchell, 1971; Yamamoto and Tanaka, 1972; Chýlek and Coakley, 1974; etc.) the interaction of the particles with terrestrial radiation was neglected because it was assumed that the IR effects were small compared with the impact of the particles on the incident solar radiation. The calculations of Rasool and Schneider (1971) which included interactions of the particles with both solar and terrestrial radiation seem to support the validity of this assumption for tropospheric aerosols. In some cases the interaction of stratospheric aerosols with terrestrial radiation can also be neglected. However, as will be demonstrated, under certain conditions stratospheric aerosols can perturb terrestrial infrared radiation more than solar radiation.

Recently, Pollack *et al.* (1976) and Harshvardhan and Cess (1976) have estimated the changes in the global climate that could be caused by an increase in stratospheric aerosols. Pollack and his collaborators

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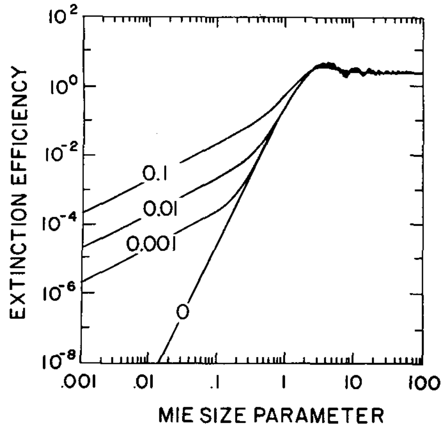

FIG. 1. Extinction efficiency factor as a function of the Mie size parameter. The real part of the index of refraction is 1.5, the imaginary parts 0, 0.001, 0.01 and 0.1.

computed the effect of a stratospheric aerosol layer on the radiative fluxes for a model of the atmosphere in which the temperature profile was specified. They inferred the response of the surface temperature to an increase in stratospheric aerosols by adjusting the atmospheric profile and then determining the value of the surface temperature that resulted in radiative equilibrium at the top of the atmosphere. By following the procedure they were unable to insure that the stratosphere and lower atmosphere would separately reach a state of radiative equilibrium as is achieved in the thermal equilibrium model described by Manabe and Wetherald (1967). Nevertheless, the work of Coakley and Schneider (1974) and Cess (1974) has demonstrated that the stratospheric temperature profile plays only a minor role compared with the tropospheric temperature profile in determining the radiative fluxes at the top of the atmosphere. As a result the estimates of surface temperature change obtained by Pollack and his colleagues are representative of those that would be obtained using a thermal equilibrium model. Harshvardhan and Cess developed a radiative energy balance model for both the stratospheric aerosol layer and the atmosphere below the layer to obtain surface temperature changes due to an increase in stratospheric aerosols. For the size distributions and optical properties they selected, both Pollack and his colleagues and Harshvardhan and Cess found that an increase in stratospheric aerosols caused lower surface temperatures. In addition, Harshvardhan and Cess suggested that stratospheric heating resulting from a large increase in the concentration of stratospheric aerosols, such as occurred after the eruption of Mt. Agung in 1963, could cause significantly higher stratospheric temperatures.

In this paper we describe a simple, radiative energy balance model that may be used to estimate the response of the global mean surface temperature to an increase in stratospheric aerosols. In the model, radiative equilibrium is maintained at both the base and the top of the stratospheric layer. Provided the tropospheric lapse rate, cloud parameters and relative humidity remain constant (these parameters are usually specified as constants in thermal equilibrium models), increases of the net radiative flux into the troposphere due to changes in the concentration of stratospheric aerosols result in warmer tropospheric temperatures. The warmer tropospheric temperatures in turn lead to increases in the upward flux of terrestrial radiation at the base of the stratosphere. The radiative energy balance model is used to derive an analytic expression for the change in the upward flux of terrestrial IR radiation at the base of the stratospheric aerosol layer caused by a specified increase in the mass concentration of the aerosol particles. The change in the upward flux of terrestrial IR radiation is used along with the results of existing climate and thermal equilibrium model calculations (Budyko, 1969; Manabe and Wetherald, 1967) to infer the aerosol-induced change in the surface temperature. The change in the upward flux is calculated as a function of the size of the aerosol particles for a range of visible and infrared optical properties of the aerosol. By computing the change in the upward flux as a function of particle size, we demonstrate the feasibility of estimating an upper limit to the response of the surface temperature for a particular increase in the mass concentration of stratospheric aerosols. Finally, the radiative energy balance model that we present separates the competing effects of the particles on the solar and terrestrial radiative fluxes. We use the results of these sample calculations to test the assumption that the major effect of the observed stratospheric particles is on the solar radiation.

2. Scattering and absorption by spherical particles

Before developing the radiative energy balance model it is well to review the general properties of the scattering and absorption of radiation by spherical particles so that we may anticipate the results obtained by using the model. Since at present there is no simple procedure for computing the scattering and absorption of radiation by a collection of particles of irregular shape, we must rely on Mie theory for the scattering of electromagnetic radiation by spherical dielectric particles to approximate the actual interaction between the stratospheric aerosol particles and the radiation field. In particular, we would like to compare the scattering and absorption properties of particles at the different wavelengths associated with solar and terrestrial radiation.

We begin by noting that the Mie size parameter of a spherical particle, given by $x=2\pi b/\lambda$ where b is the radius of the particle and λ the wavelength of the radiation, is larger for radiation at the wavelengths associated with incident solar radiation than it is at the infrared wavelengths associated with the earth's terrestrial radiation. For instance, a spherical particle having a radius of 0.3 μm, which may be a representative size for stratospheric aerosols (Friend, 1966), has a size parameter $x \approx 4$ at the solar wavelength of 0.5 μm while at 10 μm (the infrared window region of the earth's atmosphere) it has a size parameter $x \approx 0.2$.

In Fig. 1 the Mie extinction efficiency factor is plotted as a function of the size parameter for several values of n', the imaginary part of the complex index of refraction. The real part n is 1.5. The extinction efficiency of a particle is equal to the ratio of the sum of the total scattering and the absorption cross sections of the particle to its geometric cross section. The optical depth of a layer of particles is therefore proportional to the product of the extinction coefficient and the geometric cross section of the particles. As shown in Fig. 1, the extinction efficiency for small ($x \ll 1$) absorbing particles ($n' \neq 0$) is linearly proportional to the size parameter of the particles (van de Hulst, 1957). In the region of extremely small size parameters, the absorption efficiency is much larger than the scattering efficiency of the particles, as long as the imaginary refractive index is nonzero. At larger size parameters ($x \gtrsim 1$) the scattering efficiency of the particles becomes the dominant term in the extinction efficiency factor.

If we compare the extinction efficiency of a 0.3 μm particle at a wavelength of 0.5 μm with its efficiency at a wavelength of 10 μm, we find (as shown in Fig. 2) that regardless of the amount of absorption at either the visible or IR wavelength (as indicated by the range of n') the extinction efficiency of the particle for radiation at 0.5 μm is considerably larger than it is for radiation at 10 μm. Since the extinction efficiency of a given particle is proportional to the optical depth of a layer composed of such particles, it is possible to infer from Fig. 2, at least for a layer of particles with a radius of 0.3 μm, that the ratio $\tau/\tau' > 10$, where τ is the visible optical depth and τ' the infrared optical depth. Calculations for particles of similar sizes have undoubtedly contributed to the belief in the validity of the assumption that aerosols affect visible solar radiation more strongly than they affect infrared radiation.

It should be noted, however, according to Fig. 1, that the infrared extinction efficiency of an extremely small particle ($x \ll 1$) that absorbs at IR wavelengths may in fact be larger than its extinction efficiency at visible wavelengths, especially if the particle is nonabsorbing ($n'=0$) at visible wavelength and strongly absorbing ($n'>0$) at IR wavelengths. We also see in Fig. 1 that for large particles ($x \gg 1$), the visible and infrared extinction efficiencies may be of the same order of magnitude despite the difference in the size parameters of the particles at the different wavelengths. Thus, depending on the optical properties of the particles, we might expect that a stratospheric aerosol layer may have a larger effect on terrestrial IR radiation than on incident solar radiation if the layer is composed chiefly of extremely small particles, whereas if it is composed chiefly of very large particles its effects on terrestrial and solar radiation will probably be comparable.

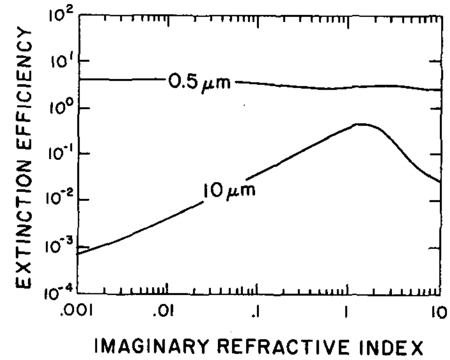

FIG. 2. Extinction efficiency factor as a function of the imaginary part of the index of refraction. The real part is taken to be 1.5. The curves are for a spherical particle having a radius of 0.3 μm. The upper curve is for a visible wavelength (0.5 μm), the lower curve an infrared wavelength (10 μm).

Although the visible and IR extinction efficiencies of a particle are useful in qualitatively establishing the relative magnitudes of the visible and IR effects of the particles in a stratospheric aerosol layer, as demonstrated in the next section, additional information is needed to estimate the relative importance of these two effects quantitatively.

3. Radiative energy balance model

We divide the atmosphere into two regions—an isothermal stratosphere and a troposphere, as illustrated in Fig. 3. The stratosphere and the earth-atmosphere system as a whole are assumed to be in radiative equilibrium. For radiative equilibrium the net downward solar radiative flux is equal to the net upward flux of terrestrial radiation at each level in the stratosphere. In particular, the condition of radiative equilibrium applied at the top of the atmosphere and at the base of the stratosphere gives

$$S_0 = F_0^+, \quad (1)$$

$$S_1 = F_1^+ - F_1^-, \quad (2)$$

where S_0 and F_0^+ are the net incoming solar and outgoing terrestrial radiative fluxes, respectively, at the top of the atmosphere and S_1 and $F_1^+ - F_1^-$ are the net solar and terrestrial radiative fluxes, respectively, at the base of the layer as illustrated in Fig. 3.

It is important at this point to emphasize the necessity of considering separately the tropospheric and stratospheric energy balance when estimating the response of the surface temperature to a change in the stratospheric aerosol layer. Previous studies of the climatic impact of aerosols have focused on the response of the radiative fluxes at the top of the atmosphere to changes in aerosol properties (e.g., Rasool and Schneider, 1971, and Chýlek and Coakley, 1974); therefore, these studies have been concerned with the energy balance of the entire earth-atmosphere system without regard to the changes in the vertical thermal

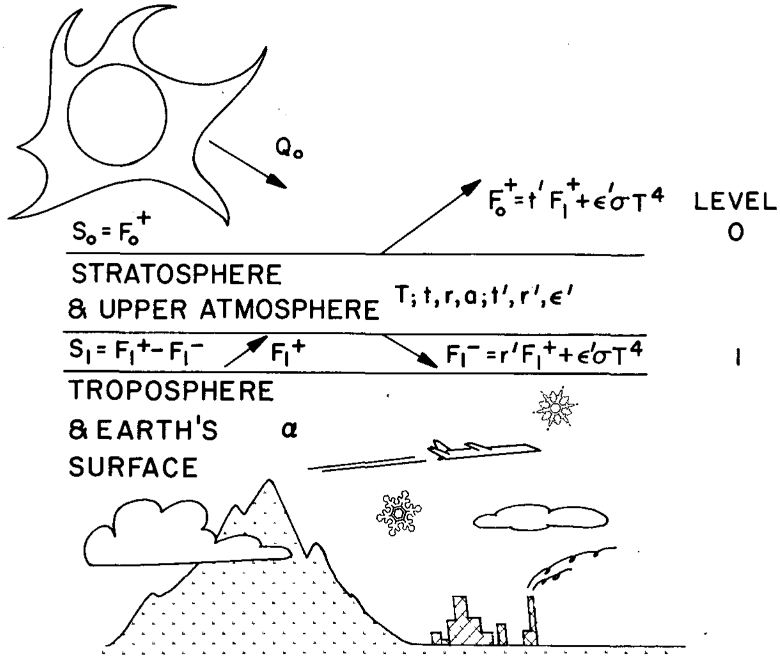

FIG. 3. Two-layer atmospheric model used to compute $\Delta F_1^+/F_1^+$, the fractional change in the upward flux of terrestrial infrared radiation at the base of the stratospheric aerosol layer. The parameters S_0 and S_1 are the net solar fluxes at levels 0 and 1 and are equal to the net fluxes of terrestrial radiation, F_0^+ and $F_1^+ - F_1^-$, at those levels when the stratosphere and the entire earth-atmosphere system are in radiative equilibrium. The parameters t and r denote the effective transmissivity and reflectivity of the stratosphere to solar radiation. The primed quantities t' and r' denote the effective transmissivity and reflectivity to terrestrial radiation. The parameter a represents the absorptivity of the stratosphere for solar radiation while ϵ' represents the layer's infrared emissivity. The temperature of the stratosphere is denoted by T. The expressions which relate F_1^- and F_0^+ to F_1^+ and T follow from the equation of radiative transfer under the assumptions that the radiation is diffuse and that the constituents of the stratosphere are gray absorbers and scatterers. The parameter α is the reflectivity of the troposphere and earth's surface and is set equal to the present value of the earth's planetary albedo. The solar constant is denoted by Q_0.

structure of the atmosphere caused by aerosols. In the case of a change in the stratospheric aerosol layer, it is possible that an increase in the absorption of solar radiation by the stratospheric aerosols would result in a lower planetary albedo and thus in a warming of the earth-atmosphere system. But, as will be shown later, the increased absorption would also cause a decrease in the rate at which energy enters the troposphere and thus result in lower tropospheric temperatures. To account for this possibility we have focused our attention in the present model on the energy balance of the troposphere as given by the radiative fluxes at the base of the stratospheric layer rather than in the energy balance of the earth-atmosphere system. As mentioned in the Introduction, we derive an expression for the fractional change in the upward flux of terrestrial radiation at the base of the stratosphere. The fractional changes in this term are indicative of changes in the energy balance of the troposphere caused by changes in the concentration of stratospheric aerosols.

Both F_0^+, the upward infrared radiative flux at the top of the atmosphere, and F_0^-, the downward infrared radiative flux at the base of the stratosphere, are related to F_1^+, the upward infrared radiative flux at the base of the stratosphere. In terms of F_1^+ they are

$$F_0^+ = t'F_1^+ + \epsilon'\sigma T^4, \tag{3}$$

$$F_1^- = r'F_1^+ + \epsilon'\sigma T^4. \tag{4}$$

Here t', r' and ϵ' are the infrared transmissivity, reflectivity and emissivity of the stratosphere; σ is the Stefan-Boltzmann constant; and T is the stratospheric temperature. To obtain (3) and (4) from the equation of radiative transfer it is necessary to use a diffusivity factor to describe the dependence of the specific intensity on zenith angle and to assume that the mean transmission of a layer composed of two separate layers is given by the product of the mean transmission of those layers. The validity of the diffuse approximation for infrared radiation has been investigated, for example, by Rodgers and Walshaw (1966). The second assumption is valid if the gases in the atmosphere are gray absorbers. Of course the gases are not gray but, as will be shown later, the absorption properties of the gases have little effect on our results for the aerosol-induced change in the surface temperature. It should be recognized that (1) and (2) apply only for the wavelength-integrated solar and infrared radiative fluxes. Consequently, t', r' and ϵ' in (3) and (4) are given by

$$t'=\frac{1}{F_1^+}\int_0^\infty d\lambda F_{1\lambda}^+ t_\lambda', \tag{5}$$

$$r'=\frac{1}{F_1^+}\int_0^\infty d\lambda F_{1\lambda}^+ r_\lambda', \tag{6}$$

$$\epsilon'=\frac{1}{\sigma T^4}\int_0^\infty d\lambda B_\lambda(T)\epsilon_\lambda', \tag{7}$$

where t_λ', r_λ' and ϵ_λ' are the monochromatic transmissivity, reflectivity and emissivity of the layer at the wavelength λ; $B_\lambda(T)$ is the Planck function at the temperature T; and $F_1^+=\int_0^\infty F_{1\lambda}^+ d\lambda$, where $F_{1\lambda}^+$ is the upward flux of radiation with wavelength λ at the base of the stratospheric layer.

Substituting the expressions for F_0^+ and F_1^- into (1) and (2) and adding gives

$$S_0+S_1=(1+t'-r')F_1^+. \tag{8}$$

We will use (8) to derive an expression for the fractional change in the upward flux of terrestrial radiation below the layer, $\Delta F_1^+/F_1^+$, and $\Delta F_1^+/F_1^+$ will in turn be related to surface temperature perturbations. Because t' for the stratosphere as defined by (3) and (5) is nearly equal to unity, $\Delta F_1^+/F_1^+$ will, to a fair approximation, depend only on the optical properties of the aerosols and will be independent of the properties of the gaseous absorbers.

Calculations of radiative fluxes for models of the earth's atmosphere may be used to demonstrate the validity of the approximation that $t'\approx 1$. We note that

$$[F_0^+ +(F_1^+ -F_1^-)]/F_1^+ =1+t'-r'. \tag{9}$$

Since $r'=0$ for a gaseous atmosphere,

$$t'=\{[F_0^+ +(F_1^+ -F_1^-)]/F_1^+\}-1. \tag{10}$$

Using Eq. (10) to compute t' based on a model for IR transfer described by Rodgers and Walshaw (1966), we arrive at a value of about 0.86 for t' associated with the region of the atmosphere above 15 km. This includes the stratospheric aerosol layer in the 15–25 km altitude interval as observed by laser radar (Grams and Fiocco, 1967) and by searchlight (Elterman *et al.*, 1973) after the eruption of Mt. Agung in 1963.

When aerosols are added to the stratosphere, the optical properties of the stratosphere change so that

$$\Delta S_0+\Delta S_1=(\Delta t'-\Delta r')F_1^+ +(1+t'-r')\Delta F_1^+, \tag{11}$$

which is obtained from (8). Thus,

$$\frac{\Delta F_1^+}{F_1^+}=\frac{\Delta S_0+\Delta S_1}{S_0+S_1}-\frac{(\Delta t'-\Delta r')}{1+t'}, \tag{12}$$

where $r'=0$ is taken for the stratosphere with no aerosols.

Next we relate $(\Delta S_0+\Delta S_1)/(S_0+S_1)$ to the changes in the visible optical properties of the stratosphere induced by the addition of aerosol particles. Taking into account the multiple reflections between the stratosphere and the lower atmosphere and earth's surface, which is assumed to be a diffusely reflecting system, S_0 and S_1 are given by

$$S_0=[1-r-\alpha t^2/(1-\alpha r)]Q_0, \tag{13}$$

$$S_1=[(1-\alpha)t/(1-\alpha r)]Q_0. \tag{14}$$

The parameters r and t are, respectively, the reflectivity and transmissivity of the layer averaged over the zenith angles of the incident solar radiation; and Q_0 is the incident flux of solar radiation at the top of the layer ($Q_0=\sigma_0/4$ is the global mean incident flux where σ_0 is the solar "constant," $\sim$1350 W m^{-2}). The albedo α is the reflectivity of the lower atmosphere and surface combined; it is taken to be equal to the planetary albedo—about 0.3 (Raschke *et al.*, 1973). It is assumed that α is independent of wavelength and that the lower atmosphere and surface reflect radiation diffusely when combined. As with r' and t', r and t are flux-weighted averages of the monochromatic reflectivity and transmissivity. They are given by

$$t=\frac{1}{Q_0}\int_0^\infty d\lambda t_\lambda Q_\lambda,$$

$$r=\frac{1}{Q_0}\int_0^\infty d\lambda r_\lambda Q_\lambda,$$

where

$$Q_0=\int_0^\infty d\lambda Q_\lambda.$$

The use of flux-weighted averages is made possible by the fact that $t\approx 1$ and $r\ll 1$ for the earth's stratosphere. The validity of these approximations may be demonstrated by using (13) and (14) to derive expressions for r, t and α in terms of S_0, S_1, and the downward flux of solar radiation at the base of the layer. Using values for the radiative fluxes numerically computed by Braslau and Dave (1972) for a model of the earth's atmosphere, we find that $t\approx 0.96$ and $r\approx 0.01$, when $\alpha\approx 0.3$ for the atmosphere above 15 km and for a 60° zenith angle (equivalent to the global mean zenith angle). With these approximations for r and t it is possible to expand the expressions for S_0 and S_1, noting that $\alpha r \ll 1$. After expanding, we obtain

$$\frac{S_0+S_1}{Q_0} \approx 1-r-\alpha t^2(1+\alpha r)+(1-\alpha)t(1+\alpha r) \quad (15)$$

$$\approx 2(1-\alpha)-2r(1-\alpha)^2-a(1-3\alpha), \quad (16)$$

where we have used the substitution $t=1-a-r$ and kept only the terms that are linear in a and r; the parameter a is the absorptivity of the layer.

We see that, with the addition of aerosols, the change in S_0+S_1 is given by

$$\frac{\Delta S_0+\Delta S_1}{Q_0} \approx -2\Delta r(1-\alpha)^2-\Delta a(1-3\alpha), \quad (17)$$

and the fractional change is given by

$$\frac{\Delta S_0+\Delta S_1}{S_0+S_1} = -\Delta r(1-\alpha)-\frac{\Delta a}{2}\frac{(1-3\alpha)}{1-\alpha}. \quad (18)$$

Once again, we have kept only terms linear in r, a, Δr and Δa.

Substituting (18) into (12) and making use of the fact that $t' \approx 1$, we obtain

$$\frac{\Delta F_1^+}{F_1^+} = \frac{\Delta r'-\Delta t'}{2}-\Delta r(1-\alpha)-\frac{\Delta a}{2}\frac{(1-3\alpha)}{1-\alpha}. \quad (19)$$

All that remains is to relate the changes in the optical properties of the stratosphere to the optical properties of the aerosols and the gases. We begin by assuming that the stratospheric aerosol optical depth will be small at all wavelengths. Elterman *et al.* (1973) found it to be about 0.02 at 0.5 μm after the eruption of Mt. Agung. It is not likely that the optical depth will be more than a few hundredths at other wavelengths. Since the optical depth is small, we may assume that radiation passing through the aerosol layer is scattered only once by the particles. Then for monochromatic radiation the transmissivity t_c of a layer containing gases with transmissivity t_g and aerosols with transmissivity t_a is given by

$$t_c = t_g t_a \quad (20)$$

$$= t_g(1-r_a-a_a), \quad (21)$$

where r_a and a_a are the reflectivity and absorptivity of the aerosol particles. Rewriting Eq. (21) we see that

$$t_c = t_g - t_g r_a - t_g a_a \quad (22)$$

$$= 1-r_g-a_g-t_g r_a - t_g a_a. \quad (23)$$

Thus the increases in the absorptivity, reflectivity and transmissivity of the layer caused by the addition of the aerosol particles are given by

$$\Delta a_c = t_g a_a, \quad (24)$$

$$\Delta r_c = t_g r_a, \quad (25)$$

$$\Delta t_c = -t_g a_a - t_g r_a. \quad (26)$$

Eqs. (24)–(26) also apply to the flux-weighted quantities if we again assume the optical properties of the gases to be independent of wavelength. Since the optical depth of the aerosols is assumed to be small, we may use a thin-atmosphere approximation to estimate global average values of a_a and r_a (Coakley and Chýlek, 1975). They are

$$a_a = 2\tau(1-\omega), \quad (27)$$

$$r_a = 2\tau\omega\beta, \quad (28)$$

where τ is the optical depth of the aerosol layer, ω the single-scattering albedo for the aerosol layer, and β the upward-scattering parameter. The parameters ω and β are related to the scattering phase function $p(\mu,\mu')$ of the aerosol by

$$\omega = \frac{1}{2}\int_{-1}^{1} d\mu p(\mu,\mu') = \frac{1}{2}\int_{-1}^{1} d\mu p(\mu,-\mu'), \quad (29)$$

$$\omega\beta = \frac{1}{2}\int_0^1 d\mu \int_0^1 d\mu' p(\mu,-\mu'). \quad (30)$$

The flux-weighted quantities are defined in the usual way by

$$\tau = \frac{1}{Q_0}\int_0^\infty d\lambda \tau_\lambda Q_\lambda, \quad (31)$$

$$\omega = \frac{1}{\tau Q_0}\int_0^\infty d\lambda \omega_\lambda \tau_\lambda Q_\lambda, \quad (32)$$

$$\omega\beta = \frac{1}{\omega\tau Q_0}\int_0^\infty d\lambda \omega_\lambda \beta_\lambda \tau_\lambda Q_\lambda. \quad (33)$$

Substituting the above expressions for Δr [Eq. (28)] and Δa [Eq. (27)] along with similar expressions for $\Delta r'$ and $\Delta t'$ ($\Delta t = -\Delta r - \Delta a$) in (19) and making use of the fact that both t' and t are near unity gives

$$\frac{\Delta F_1^+}{F_1^+} \approx \tau'[1-\omega'+2\omega'\beta'] - \tau\left[(1-\omega)\frac{(1-3\alpha)}{(1-\alpha)}+2\omega\beta(1-\alpha)\right]. \quad (34)$$

Finally, it is necessary to relate $\Delta F_1^+/F_1^+$ to a change in the global mean surface temperature. We may obtain an approximate estimate from the work of Budyko (1969) and Manabe and Wetherald (1967). Budyko estimated that a 1.6 K decrease in surface temperature resulted in a 1% decrease in the upward flux of terrestrial radiation at the top of the atmosphere. Similarly, Manabe and Wetherald found in their thermal equilibrium model of the earth's atmosphere that a 1% decrease in the upward flux at the top of the atmosphere was caused by about a 1 K decrease in surface temperature. Thus, for the purpose of estimating the surface temperature response, we will assume that a 1% change in F_1^+ at the base of the stratosphere corresponds to a 1 K change of like sign in global mean surface temperature. To arrive at this relationship we have neglected any possible connection between the thermal structure of the stratosphere and the flux of thermal radiation leaving the top of the atmosphere.

It should be remembered that the relationship between the change in surface temperature and $\Delta F_1^+/F_1^+$ is only a rough approximation. It is based in part on model results that do not account for possible cloud feedback mechanisms as described by Schneider (1972) and Cess (1974) and albedo-temperature feedback mechanisms as modeled by Budyko (1969) and Sellers (1969), as well as other yet-to-be-determined climatic feedbacks. Such feedback mechanisms could alter the optical properties of the lower atmosphere which could in turn lead to a change in the net radiative flux received by the troposphere and consequently to altered tropospheric temperatures. The changes in tropospheric temperature would then be reflected in the values of $\Delta F_1^+/F_1^+$. Thus, because of our inability to adequately determine the magnitude of various feedback mechanisms, the relationship between $\Delta F_1^+/F_1^+$ and surface temperature perturbations remains uncertain. With this in mind, we have ignored any possible link between stratospheric temperatures and the upward flux of thermal radiation at the top of the atmosphere noting that this link is in any case small compared to the link between the outgoing flux and surface temperature (Coakley and Schneider, 1974; Cess, 1974). By using our approximate relationship between $\Delta F_1^+/F_1^+$ and the change in surface temperature, we may use (34) to compute $\Delta F_1^+/F_1^+$ attributable to an increase in stratospheric aerosols, and then use the computed value to estimate the surface temperature response.

In Eq. (34) we have separated the effects of the aerosol on the solar and terrestrial radiative fluxes. The term in the first brackets on the right-hand side constitutes the IR contribution and we see (as speculated in the Introduction) that this contributes to a warming at the surface. The remaining term is associated with the effects of the aerosols on the incident solar radiation. Since the planetary albedo $\alpha \approx 0.3$, this term contributes to surface cooling. On the other hand, it should be recognized that if the planetary albedo were sufficiently large (which may be the case during glacial periods) an aerosol layer that strongly absorbs solar radiation might also contribute to surface warming by its effects on the incident solar radiation.

Note that a procedure similar to the one used to determine $\Delta F_1^+/F_1^+$ could, in principle, be used to determine the fractional change in the stratospheric temperature. Subtracting (2) from (1) and substituting the expressions for F_0^- and F_0^+, we obtain

$$S_0 - S_1 = 2\epsilon'\sigma T^4 - (1-r'-t')F_1^+. \tag{35}$$

If we assume the gases and aerosols are gray absorbers, then $1-r'-t'=\epsilon'$ and (35) becomes

$$\sigma T^4 = \frac{S_0 - S_1}{2\epsilon'} + \frac{F_1^+}{2} \tag{36}$$

$$= \frac{S_0 - S_1}{2\epsilon'} + \frac{S_0 + S_1}{2(1-r'-\epsilon'/2)}. \tag{37}$$

Substituting for S_0 and S_1 and making use of our previous assumptions along with the additional fact that $\epsilon' \ll 1$, we find that

$$\frac{\Delta T^4}{T^4} \approx \left(\frac{\Delta a}{a} - \frac{\Delta\epsilon'}{\epsilon'}\right)\left[\frac{a(1+\alpha)}{a(1+\alpha)+\epsilon'(1-\alpha)}\right]. \tag{38}$$

Since the factor in the brackets is positive, an increase in stratospheric aerosols that are nonabsorbing at the wavelengths of solar radiation ($\Delta a = 0$), but which do absorb in the infrared, will lead to cooler stratospheric temperatures for the case of gray absorbers. This is consistent with the findings of Harshvardhan and Cess (1976). It should be noted, however, that the earth's atmosphere and the aerosol particles are not gray and, therefore, in general $\epsilon' \neq 1-r'-t'$. Harshvardhan and Cess showed, in fact, that if the aerosol were sufficiently nongray an increase in stratospheric temperatures could result from an increase in stratospheric aerosols.

We note that the present model is probably inadequate for estimating the changes in *stratospheric* temperatures due to an increase in stratospheric aerosols because we have considered the entire upper part of the atmosphere to be in one layer. Thus, $\Delta a/a$ and $\Delta\epsilon'/\epsilon'$ are likely to be very small since the gases will probably dominate the absorption within the layer. We would have to divide the stratosphere into at least two layers—one extending over the entire aerosol layer and another without aerosols—to obtain a better estimate of the response of the layer temperature to an increase in stratospheric aerosols. The addition of a third layer would complicate the model significantly. However, since the results for $\Delta F_1^+/F_1^+$ are relatively insensitive to the optical properties of the gases, the addition of a third layer would not alter our findings for the changes in the *surface* temperature, which is of primary concern to us here.

Before proceeding to numerical calculations of $\Delta F_1^+/F_1^+$, we compare the results presented in (34) to the results obtained from the argument that an increase in stratospheric aerosols will result in a decrease in the downward flux of solar radiation to the troposphere and

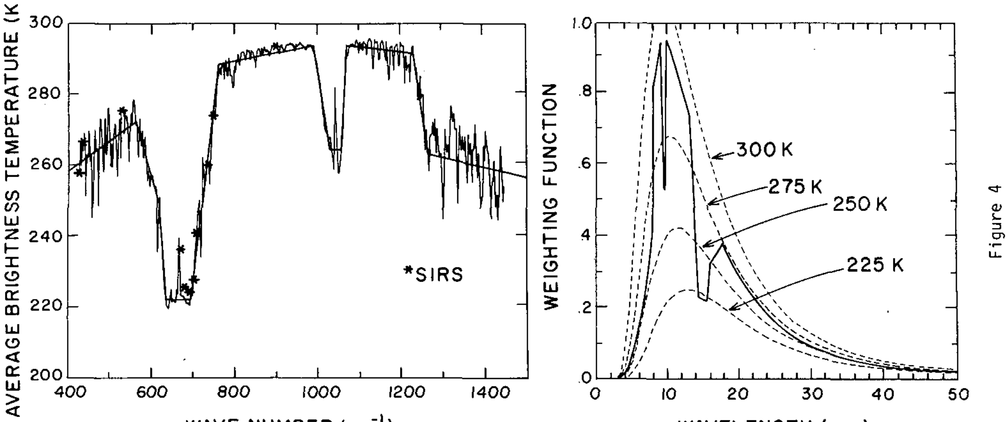

FIG. 4. Brightness temperatures obtained from IRIS and SIRS satellite observations reported by Kunde *et al.* (1974) and weighting function used to compute effective optical properties of the stratospheric aerosol layer for terrestrial radiation.

thus can be approximated by an equivalent decrease in the solar constant. This is the effect of the argument put forward by Mitchell (1971), and it has been used by others (Coakley and Schneider, 1974). In this approximation,

$$\frac{\Delta F_{\mathrm{I}}^{+}}{F_{\mathrm{I}}^{+}} \approx \frac{\Delta F_D}{F_D}, \tag{39}$$

where the fractional change in the downward flux of solar radiation below the aerosol layer is denoted by $\Delta F_D/F_D$. Using the same approximations that we used previously, we arrive at

$$\frac{\Delta F_D}{F_D} \approx -2\tau[1-\omega+(1-\alpha)\omega\beta]. \tag{40}$$

Thus, in the absence of infrared effects and for the case in which the aerosols are nonabsorbing at the wavelengths of the incident solar radiation (i.e., $\omega=1$), we see that (34) reduces to (40). On the other hand, we might expect that simulating an increase in stratospheric aerosols by an equivalent decrease in the solar constant would lead to a poor estimation of the surface temperature response if the aerosol strongly absorbs solar radiation. As mentioned earlier, Eq. (34) suggests that the effects of the stratospheric aerosol layer on the incident solar radiation can also contribute to surface warming, provided the planetary albedo and absorptivity of the aerosol are sufficiently large. Because the earlier arguments did not consider the effect of the aerosols on IR radiation, they indicated that only surface cooling would result from an increase in the concentration of stratospheric aerosols, as indicated in (40).

In the following section we use (34) to compute $\Delta F_{\mathrm{I}}^{+}/F_{\mathrm{I}}^{+}$ for spherical particles and for a range of visible and IR optical properties.

4. Numerical results

For our numerical examples we chose to specify the amount of aerosols in terms of a mass concentration measured in micrograms per cubic meter ($\mu g\ m^{-3}$). This is often the unit used to measure the concentration of trace species in the atmosphere.

In the computations of $\Delta F_{\mathrm{I}}^{+}/F_{\mathrm{I}}^{+}$, we use the flux-weighted averages of the optical properties of the aerosols as defined by Eqs. (31)–(33). The spectral flux-weighting function used for the terrestrial radiation is given in Fig. 4. It was obtained from IRIS satellite measurements as described by Kunde *et al.* (1974) and, although its use may not be entirely consistent with our globally averaged model, we would anticipate little effect on our calculations if other data were used as a spectral weighting function. For the solar spectral flux we used the wavelength distribution of a 6000 K blackbody.

As mentioned earlier, the particles are assumed to be spherical and Mie theory is used to compute the appropriate scattering and absorption cross sections. One index of refraction is used for solar radiation and a second for terrestrial radiation. The indices used in these calculations are 1.5–0.0i and 1.5–0.01i for solar radiation, and 1.5–0.1i and 1.5–0.5i for terrestrial radiation. These indices were chosen because the sulfates that apparently constitute the stratospheric aerosols (Cadle, 1972; Cadle and Grams, 1975) seem to be nonabsorbing at visible wavelengths and reasonably strong absorbers at IR wavelengths (Neumann, 1973).

In Fig. 5 we show the computed values of $\Delta F_{\mathrm{I}}^{+}/F_{\mathrm{I}}^{+}$ based on Eq. (34) as a function of the radius of the spherical particles. The results are for four combinations of the visible and IR indices of refraction and are for a mass concentration of 1 $\mu g\ m^{-3}$ of particles in a 1 km thick layer. This particular value of the particle concentration was chosen as typical. The effect of any other concentration may be obtained by noting that, according to (34), $\Delta F_1^+/F_1^+$ is linearly proportional to the particle concentration. Therefore, the results shown in Fig. 5 may be readily scaled to any other realistic values of the mass concentration and layer thickness of a stratospheric aerosol layer. The mass density of the particles is assumed to be 2 g cm^{-3}.

We plot $\Delta F_1^+/F_1^+$ as a function of the particle radius for two reasons. First, the size distribution of the stratospheric aerosols is uncertain, as shown by the variety of results obtained from measurements of the stratospheric aerosol size distributions (Junge, 1963; Friend, 1966; Remsberg, 1973; Cadle and Grams, 1975; among others). Furthermore, there is no reason to believe that the size distribution of particles in the stratosphere will remain constant regardless of the nature of their source. Second, after $\Delta F_1^+/F_1^+$ has been computed as a function of the particle radius, the results may later be used to infer the influence of a particular size distribution of particles on $\Delta F_1^+/F_1^+$. This is readily done by weighting the single-size results by the particular size distribution.

In Fig. 5 the infrared and visible contributions to $\Delta F_1^+/F_1^+$ are plotted separately so that a comparison may be made of their relative magnitudes. We note that the influence of the stratospheric aerosol layer on the incident solar radiation is not very sensitive to the

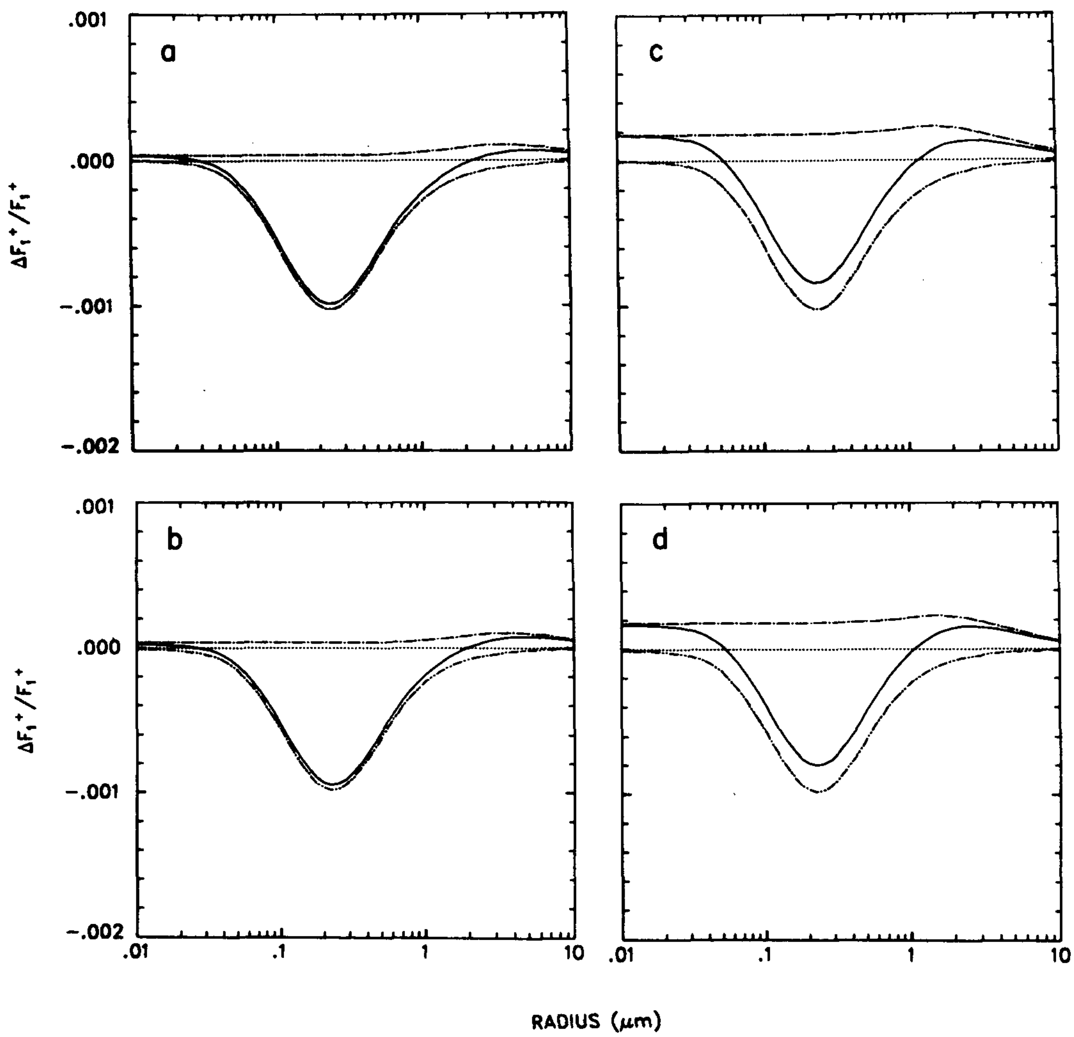

FIG. 5. Fractional change in upward flux of terrestrial radiation at the base of the stratospheric aerosol layer computed using Eq. (34) as a function of the radius of the aerosol particle. The calculations are for a mass concentration of 1 μg m^{-3} in a 1 km thick layer. The density of the aerosol particles is taken to be 2.0 gm cm^{-3}. The solid curve is the value of $\Delta F_1^+/F_1^+$; the dashed-dot curve is the contribution to $\Delta F_1^+/F_1^+$ due to the effects of the particles on terrestrial radiation, the primed term in (34); the dashed-double-dot curve is the contribution due to the effects of the particles on solar radiation, the unprimed term in (34). The dotted line indicates $\Delta F_1^+/F_1^+=0$. The indices of refraction are as follows: in (a) the index for solar radiation is 1.5–0.0*i* and for terrestrial radiation 1.5–0.1*i*; (b) 1.5–0.01*i* and 1.5–0.1*i*; (c) 1.5–0.0*i* and 1.5–0.5*i*; (d) 1.5–0.01*i* and 1.5–0.5*i*.

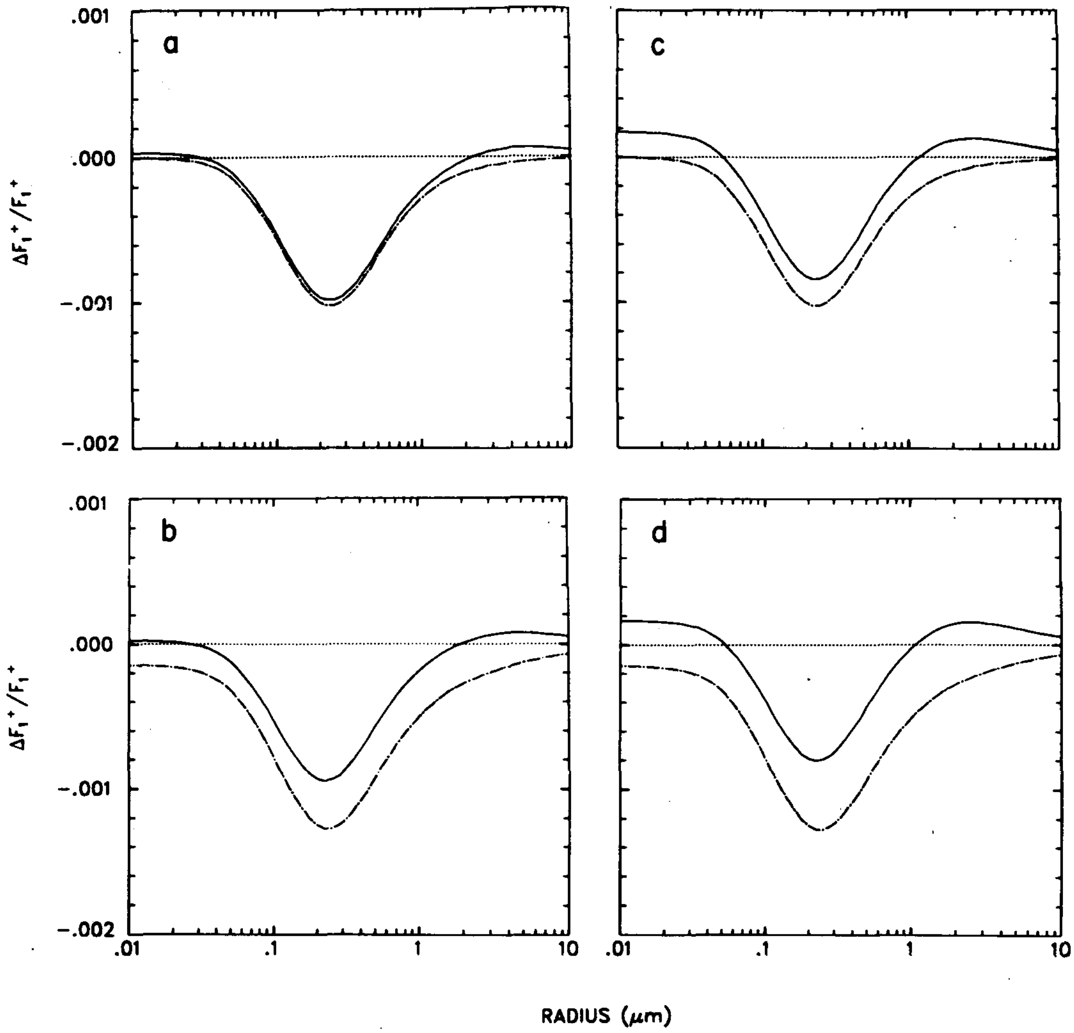

FIG. 6. Comparison of the values of $\Delta F_\mathrm{I}^+/F_\mathrm{I}^+$ calculated using the present model, the solid curve derived from Eq. (34), and those calculated assuming an equivalent reduction in the solar constant, the dashed-dot curve derived from (40). The mass density of aerosols and the indices of refraction are the same as in Fig. 5.

choice of the imaginary index of refraction for the solar spectrum, whereas large changes in the effect of the aerosol layer on terrestrial radiation follow changes in the imaginary index of refraction for the IR spectrum. This behavior is similar to that shown in Fig. 2 in terms of the extinction efficiency for the 0.3 μm particle.

As suggested earlier, the IR contribution becomes the dominant contribution for both small particles with radii on the order of a few hundredths micrometers or smaller and large particles with radii $\gtrsim 1\ \mu$m. Particles of these sizes in the stratosphere are likely to contribute to surface warming. On the other hand, particles in the intermediate size range will contribute to cooling. Finally, Fig. 5 shows that the effect of particles with radii between 0.1 and 1.0 μm on the incident solar radiation can greatly outweigh their effect on the terrestrial infrared radiation.

In Fig. 6, a comparison is made between the calculated values of $\Delta F_\mathrm{I}^+/F_\mathrm{I}^+$ using Eq. (34) and the values obtained by simulating the effect of the aerosols by making an equivalent reduction in the solar constant as computed using (40). As noted in the last section, the results are comparable when the absorption of solar radiation is small ($n'=0$) and when the IR effects are small compared with the visible effects of the aerosol, as shown for example in Fig. 6a for particles with radii between 0.1 and 1.0 μm. However, it is seen that when either the absorption of solar radiation by the particles become appreciable ($n'=0.01$) or the effect of the particles on terrestrial radiation cannot be neglected, estimating the aerosol-induced change in F_I^+ by equating it to the effect of an equivalent reduction in the solar constant as given by (40) can lead to large errors, even with regard to the sign of the changes.

For the sake of comparison we have also computed $\Delta F_\mathrm{I}^+/F_\mathrm{I}^+$ from (34) but using the optical properties of the aerosols at wavelengths of 0.5 and 10 μm instead of the flux-weighted optical properties averaged over

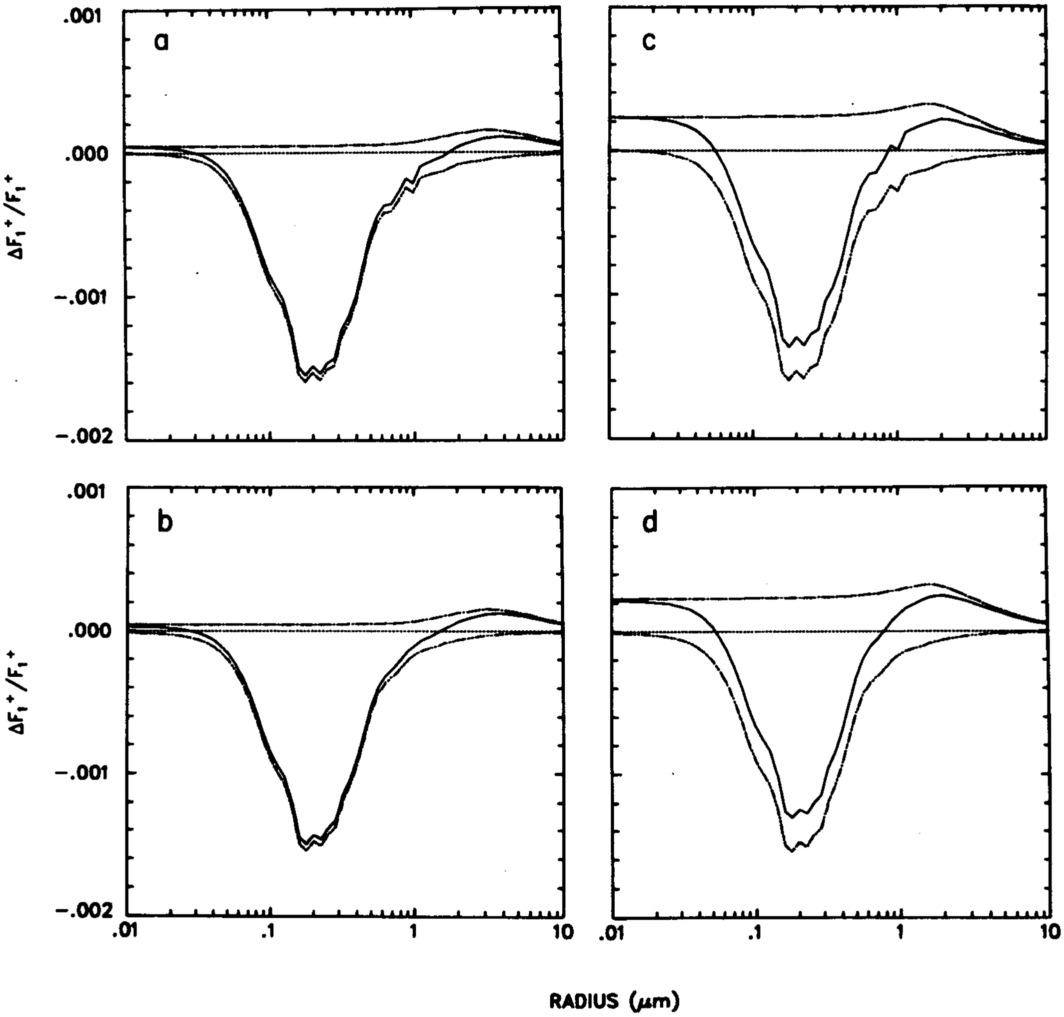

FIG. 7. Calculated values of $\Delta F_{\rm I}^+/F_{\rm I}^+$ taking 0.5 μm as an effective wavelength for solar radiation and 10 μm as an effective wavelength for terrestrial radiation. The indices of refraction, the aerosol mass concentration and the meaning of the curves are the same as in Fig. 5.

wavelengths. The results of these calculations are shown in Fig. 7. Although the results are qualitatively similar to those shown in Fig. 5, we see that the magnitude of the monochromatic value of $\Delta F_{\rm I}^+/F_{\rm I}^+$ can differ greatly from the flux-weighted value. This suggests that for realistic estimates of the magnitude of the effect of aerosols on the earth's radiation balance, the spectral properties of the absorption and scattering of both the aerosols and the gases may be important. The degree to which spectral data are necessary for quantitative estimates should be studied further.

Finally, we note one further advantage of plotting $\Delta F_{\rm I}^+/F_{\rm I}^+$ as a function of the radius of the particle. From Fig. 5 we see that it is possible to estimate the maximum possible response of $\Delta F_{\rm I}^+/F_{\rm I}^+$ that could occur for a given mass concentration of particles. Thus, if the relationship between $\Delta F_{\rm I}^+/F_{\rm I}^+$ and the accompanying change in the global mean surface temperature were well known, it would also be possible to estimate an upper limit to the surface temperature change that would occur as a result of an increase in the concentration of stratospheric aerosols. For example, from Fig. 5d we see that $\Delta F_{\rm I}^+/F_{\rm I\,max}^+ \approx -0.0008$. Using our relationship between $\Delta F_{\rm I}^+/F_{\rm I}^+$ and ΔT_s, the change in the global mean surface temperature (K), we may parameterize the results shown in Fig. 5d by

$$\Delta T_{s\ \rm max} \approx -0.8S, \qquad (41)$$

where S is now the increase in the mass concentration of stratospheric aerosols measured in micrograms per cubic meter for a 10 km thick layer. Thus, we would expect that the maximum surface temperature response attributable to the increase in stratospheric aerosols after the eruption of Mt. Agung for which $S \approx 1.0\ \mu{\rm g\ m^{-3}}$ would be $T_{s\ \rm max} \approx -0.8$ K.

That such a large response in the global mean surface temperature due to an increase in stratospheric aerosols has not been observed could be for any of a number of reasons. The mass concentration and aerosol optical properties that we have chosen may not in fact be representative of a global average stratospheric aerosol layer. It should also be remembered that the expression for $\Delta F_1^+/F_1^+$ is derived by assuming that a steady-state equilibrium exists for the earth-atmosphere system. The response time of the system may, however, be quite large due to the large thermal inertia of the oceans. In addition, the relationship between $\Delta F_1^+/F_1^+$ and changes in the surface temperature is not well known because of the uncertainties brought about by possible climate feedback mechanisms. Estimates made using the present model are useful only in that they approximate the change in the global mean equilibrium surface temperature under the conditions that the feedback mechanisms which might affect the optical properties of the lower atmosphere are not included. Under these conditions such models as the one presented here are useful for estimating the relative magnitude of two competing influences. Here we have compared the relative influence of the visible and IR optical properties of stratospheric aerosols on the surface temperature.

5. Conclusions

We have developed a simple radiative energy balance model for the earth-atmosphere system. We have used the model to determine $\Delta F_1^+/F_1^+$, the fractional change in the upward flux of terrestrial IR radiation at the base of the stratosphere that could be caused by an increase in the concentration of stratospheric aerosols. Under the assumption that the gases in the stratosphere could be treated as gray absorbers, it was shown that $\Delta F_1^+/F_1^+$ depended only on the optical properties of the aerosol particles. In part because of the assumption of gray absorbers that was used for the gases and aerosols and in part because only a single atmospheric layer was used to describe the atmosphere above the troposphere, the model is probably inadequate for approximating the change of the atmospheric temperature in the vicinity of the aerosol layer. Although these approximations make the model inadequate for the purpose of estimating the response of the local temperature to an increase in aerosols, they have little effect on the estimate of $\Delta F_1^+/F_1^+$ and therefore on the response of the surface temperature to such an increase.

We used the results of climate models and thermal energy balance calculations for the earth's atmosphere to relate $\Delta F_1^+/F_1^+$ to changes in the equilibrium global mean surface temperature. We estimated from existing climate model results that a 1% decrease in F_1^+ was accompanied by about a 1 K decrease in the surface temperature. Thus, the results we obtained for $\Delta F_1^+/F_1^+$ could be used to provide an estimate of the change in the equilibrium global mean surface temperature under the condition that the characteristics determining the optical properties of the lower atmosphere (such as cloudiness and surface albedo) are relatively independent of surface temperature.

To estimate $\Delta F_1^+/F_1^+$, the stratospheric aerosols were assumed to be spherical particles. The results obtained for $\Delta F_1^+/F_1^+$ for a range of the visible and IR indices of refraction of the aerosols have been plotted as a function of the radius of the particles for a given mass concentration of the stratospheric aerosols. This was done to demonstrate the feasibility of estimating the maximum possible response of the surface temperature for a specific mass of particulate matter in the stratosphere. The maximum response for a given mass of aerosol particles occurs when the aerosols are all of the same size, having a radius of about 0.2 μm. The actual response is likely to be significantly less than the maximum possible response because 1) it is highly unlikely that the stratospheric aerosols would form a monodispersed system with all particles having that radius and 2) the slow response time of the earth-atmosphere system is likely to have a moderating effect on the surface temperature change.

Using the relationship between $\Delta F_1^+/F_1^+$ and the change in the equilibrium global mean surface temperature, we estimate that the maximum possible surface temperature response attributable to a volcanic eruption of the magnitude of Agung is a cooling of 0.8 K. We note, based on our results for $\Delta F_1^+/F_1^+$, that because most of the mass of the stratospheric aerosols appears to be concentrated in particles with radii between 0.1 and 1 μm, the most probable contribution of present-day stratospheric aerosols is one of cooling at the surface. The possibility that present-day stratospheric aerosols contribute to surface warming seems rather unlikely considering the results of the calculations presented here. We also note that, especially for particles outside of the size range 0.1–1.0 μm, serious errors may ensue if the effect of the stratospheric aerosol layer on the global mean surface temperature is approximated by an equivalent change in the solar constant.

Finally, it should be recognized that the above conclusions are independent of the specific details of the relationship between $\Delta F_1^+/F_1^+$ and the global mean surface temperature. We note that the same conclusions would have been drawn provided that $\Delta F_1^+/F_1^+$ is only proportional to the change in the surface temperature and that the constant of proportionality is positive. We might expect that for sufficiently small deviations from the mean, $\Delta F_1^+/F_1^+$ would, in fact, be linearly proportional to the change in surface temperature. Furthermore, satellite observations such as those described by Raschke *et al.* (1973) suggest that the constant of proportionality is indeed positive, at least for the hemispheric mean surface temperature for time scales on the order of seasonal changes. Based on this evidence we conclude, as we did earlier, that the only probable surface temperature response due to an increase in stratospheric aerosols similar to those now observed is likely to be one of cooling.

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