

Stratospheric aerosols: Effect upon atmospheric temperature and global climate

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ABSTRACT

A significant fraction of the aerosol population of the stratosphere is believed to consist of sulfate, presumably in the form of sulfuric acid. An increase in stratospheric aerosols could modify global climate by augmenting reflection of sunlight as well as enhancing the atmospheric greenhouse, two competing mechanisms with regard to changing the global surface temperature. Assuming that the aerosols consist of supercooled 75 % aqueous sulfuric acid, we present a first-order estimate as to the effect of aerosol concentration upon global surface temperature. The model calculations illustrate that the increase in reflected sunlight constitutes the dominant contribution by aerosols; the normal aerosol concentration reduces the global surface temperature by roughly 0.7 K, and a doubling of concentration would provide a further decrease by the same amount. Increased aerosol concentration further results in heating of the stratosphere through absorption of infrared radiation emitted by the earth-atmosphere system. The model indicates that stratospheric temperature at 20 km could be raised by as much as 9 K, consistent with Southern Hemisphere observations following the eruption of Mt. Agung.

Introduction

There has been considerable recent concern that atmospheric aerosols, as scatterers, absorbers and emitters of radiation, might significantly alter the energy balance of the earth-atmosphere system, producing a subsequent change in our global climate. In a broad sense such aerosols may be classified as either tropospheric or stratospheric; tropospheric aerosols have a residence time usually less than a few weeks, whereas for stratospheric aerosols it is on the order of several years. Volcanic activity constitutes a major source of stratospheric aerosols, but as a consequence of their long residence time it has been suggested that high-flying aircraft and industrial activity could lead to an increasing stratospheric aerosol concentration.

There have been several recent investigations as to the effect of aerosol build up upon global climate, for example the work of Rasool & Schneider (1971), Yamamoto & Tanaka (1972), Reck (1974), and Wang & Domoto (1974), to name but a few. Such investigations have,

however, been concerned with the role of tropospheric aerosols, while the possible influence of stratospheric aerosols upon global climate requires further investigation.

Stratospheric aerosols can modify global climate in one of two ways. Reflection of solar radiation enhances the planetary albedo, reducing the global surface temperature, whereas the infrared opacity of the aerosol layer augments the greenhouse effect and increases the surface temperature. There has been considerable debate as to which of these two competing mechanisms dominates, although recently Coakley & Grams (1975) have presented a simple but convincing argument that the albedo modification is most important, so that an increase in aerosol concentration would produce a global cooling trend.

The purpose of the present study is to formulate a first-order analysis describing the magnitude of the reduction in global surface temperature as a function of aerosol concentration. Associated with this is an estimate of the corresponding change of stratospheric temperature in the vicinity of the aerosol layer.

Aerosol model

Composition measurements of stratospheric aerosols indicate that sulfate, most likely in the form of sulfuric acid, is the single most abundant chemical component (Lazrus et al., 1971; Rosen, 1971; Castleman, 1974). Vapor pressure calculations illustrate that sulfuric acid droplets should be stable under stratospheric conditions (Toon & Pollack, 1973). Rosen (1971) has shown that an aerosol composition of 75% sulfuric acid could explain observed aerosol boiling points for material sampled from the stratosphere. Some sulfate is believed to exist as ammonium sulfate, but this probably forms less than ten percent of the total aerosol content (Lazrus et al., 1971).

Although the existence of HNO_3 has been ruled out under stratospheric conditions (Toon & Pollack, 1973), it could exist incorporated with sulfuric acid (Remsberg, 1973; Castleman, 1974). The vapor pressure calculations and boiling point data give credence to the possibility that 75% sulfuric acid forms the bulk of stratospheric aerosols; at stratospheric temperatures it would appear either as a supercooled liquid or in solid form. The degree of supercooling required (35 K) is not inconsistent with supercooling in water clouds, whereas if the aerosol actually is present as a solid, Remsberg (1973) suggests that the optical properties of the solid will not differ substantially from those for the liquid. From these considerations we assume that the stratospheric aerosol consists entirely of 75% aqueous sulfuric acid in liquid form.

The size distribution adopted in the present study is that due to Remsberg (1973), for which

$$\frac{dN}{d(\log r)} = 10^{-0.474} r^{-3.82}, \quad 0.3 \leq r \leq 1 \mu\text{m} \quad (1a)$$

$$\frac{dN}{d(\log r)} = 10^{+3.72} r^{+4.19}, \quad 0.1 \leq r \leq 0.3 \mu\text{m} \quad (1b)$$

$$\frac{dN}{d(\log r)} = 10^{+4}, \quad 0.03 \leq r \leq 0.05 \mu\text{m} \quad (1c)$$

where dN/dr is the number of particles per cubic centimeter with radii between r and $r + dr$.

As pointed out by Remsberg (1973), the mass density of sulfuric acid particles corresponding to eq. (1) is highly uncertain (see

also Castleman, 1974). This will have no effect upon a global energy balance, however, since we will express the radiative properties of the aerosol layer in terms of a measured visible optical thickness rather than a sulfuric acid mass density. The mass density will, however, enter into our estimate of stratospheric temperature, such that this calculation must be regarded strictly as qualitative.

We further assume that the aerosol layer is centered at 20 km, and for the same reasons as discussed above this will influence only our estimate of stratospheric temperature. The turbidity profiles of Elterman et al. (1973) indicate that the normal stratosphere probably contains two aerosol layers centered at 15.6 km and 19.3 km, while the enhanced aerosol concentration following the Mount Agung eruption more closely resembled a single layer centered at roughly 20 km. This latter result is consistent with the observed increase in stratospheric temperature following the Agung eruption (Newell, 1970), which was greater at 19.5 km than at lower levels.

Radiative properties of the aerosol

The aerosol layer both absorbs and reflects incident solar radiation, while it absorbs, reflects and emits at infrared wavelengths. A prerequisite to the inclusion of the aerosol layer into a global energy balance is the evaluation of appropriate radiation properties for both solar and infrared wavelengths. Following Deirmendjian (1969), the spectral extinction coefficient β , applicable for both solar and infrared radiation, may be expressed as

$$\beta(\lambda) = \pi \int_{r_1}^{r_2} r^2 Q_{\text{ext}}(r, \lambda, m) \frac{dN(r)}{dr} dr \quad (2)$$

where r_1 and r_2 represent the radii of the smallest and largest particles, respectively, $Q_{\text{ext}}(r, \lambda, m)$ is the efficiency factor which relates the geometrical cross-sectional area of the particle to the optically effective cross section for extinction, while λ is wave length, $m = \nu - i\kappa$ the complex index of refraction, and dN/dr the particle size distribution as given by eq. (1).

1. Solar radiation

We first consider the evaluation of β appropriate to solar wavelengths (β_{vis}), in addition

to an estimate of the magnitude of solar absorption by the aerosol layer and a determination of the aerosol reflectivity. From the index of refraction measurements of Palmer & Williams (1975) for 75% H_2SO_4 , $\nu = 1.43$ and $\kappa = 0$ (10^{-7}). The value for ν actually refers to $\lambda = 0.55 \mu\text{m}$, but ν is a very weak function of λ for solar wavelengths; hence we assume a constant value. Although the refractive index measurements refer to 300 K, these should suffice for present purposes. The variation of κ with λ is of no consequence at solar wavelengths, since the small value of κ allows us to neglect the absorption contribution to total extinction. This will be justified shortly.

With absorption neglected, we may employ Q_{ext} for nonabsorbing spheres within eq. (2) to evaluate β_{vis} . Integration over the three ranges given by eq. (1), and computing Q_{ext} from Mie theory, gives $\beta_{\text{vis}} = 8.8 \times 10^{-3} \text{ km}^{-1}$. The visible absorption coefficient may also be computed using eq. (2) with Q_{abs} replacing Q_{ext} . For $\kappa < 1$ an asymptotic expression for Q_{abs} applies (Van de Hulst, p. 181, 1957), which is sufficiently accurate for $\nu = 1.43$ when $\kappa = 0$ (10^{-7}) (Plass, 1966). From this it readily follows that

$$Q_{\text{abs}} \simeq 4\pi x$$

where $x = 2\pi r/\lambda$ is the size parameter. Use of the above in eq. (2), and integrating over the size distribution given by eq. (1), yields a visible absorption coefficient which is of 0 (10^{-6}) compared to β_{vis} , justifying our use of Mie functions for nonabsorbing spheres in computing β_{vis} .

The same conclusion applies to an energy balance for the aerosol layer. With $(1 - \tilde{\omega}) = 10^{-6}$, where $\tilde{\omega}$ is the single scattering albedo for the polydispersion, and employing the infrared absorptivity of the following section, we find that solar absorption by the aerosol layer is more than four orders of magnitude less than infrared absorption. Our radiative model for the aerosol layer at solar wavelengths thus consists of a gray reflecting layer.

With $\beta_{\text{vis}} = 8.8 \times 10^{-3} \text{ km}^{-1}$, we may estimate a scale dimension L of the aerosol layer for use in determining the temperature at the location of the layer. In terms of this scale dimension, the visible optical thickness of the aerosol layer may be expressed as $\tau_{\text{vis}} = \beta_{\text{vis}} L$. We take

$\tau_{\text{vis}} = 0.02$ for the normal stratosphere (Elterman et al., 1973), although we caution that this is a very uncertain value. The aerosol scale dimension is thus $L = 2.3 \text{ km}$. From the aerosol turbidity profiles of Elterman (1973), this seems to be a reasonable value for enhanced aerosol concentrations, but it would appear to be an underestimate for the normal stratosphere. Nevertheless it will suffice for qualitative considerations of stratosphere temperature.

It remains to determine the solar reflectivity of the aerosol layer. The optically thin limit is appropriate for present purposes, for which the reflectivity R is

$$R = 2\tilde{\omega}b\tau_{\text{vis}} \quad (3)$$

where b is the backscatter parameter defined by Coakley & Chylek (1975). This parameter is related to the scattering phase functions by the relation

$$b = \frac{1}{4\pi} \int_0^\pi [P_1(\theta) + P_2(\theta)] \theta \sin \theta d\theta \quad (4)$$

where $P_1(\theta)$ and $P_2(\theta)$ are the normalized phase functions for the polydispersion (Deirmendjian, p. 75, 1969). Evaluation of $P_1(\theta)$ and $P_2(\theta)$ for the size distribution given by eq. (1), and use of eqs. (3) and (4) with $\tilde{\omega} = 1.0$, yields

$$R = 0.37 \tau_{\text{vis}} \quad (5)$$

2. Infrared radiation

For infrared wavelengths, $x = 2\pi r/\lambda < 1$, and series expansions (Penndorf, 1962) may be employed to evaluate Q_{abs} and Q_{sca} . The evaluation of the infrared absorption coefficient, $\beta_{\text{IR}}(\lambda)$, then follows from eq. (1) and (2) together with Penndorf's expression for Q_{abs} . The real and imaginary indices of refraction are again taken from Palmer & Williams (1975). The same procedure has been used to determine the single scattering albedo, and we find that $\tilde{\omega}(\lambda) < 0.1$ for $\lambda > 5 \mu\text{m}$, such that scattering may be neglected in performing an energy balance on the aerosol layer. This is consistent with the conclusions of Remsberg (1973), and Coakley & Grams (1975).

Recalling that our basic aerosol parameter is τ_{vis} , then the spectral infrared optical thickness is expressed by

$$\tau_{\text{IR}}(\lambda) = (\beta_{\text{IR}}/\beta_{\text{vis}})\tau_{\text{vis}} \quad (6)$$

with $\beta_{\text{vis}} = 8.8 \times 10^{-3} \text{ km}^{-1}$. Although β_{vis} and β_{IR} individually depend upon the aerosol mass density, which is an uncertain quantity, the ratio $\beta_{\text{IR}}/\beta_{\text{vis}}$ is independent of mass density, providing the size distribution is independent of mass density.

The slab emissivity of the aerosol layer is defined as

$$\varepsilon = \frac{1}{\sigma T^4} \int_0^\infty e_\lambda(T) [1 - 2E_3(\tau_{\text{IR}})] d\lambda \quad (7)$$

where $e_\lambda(T)$ is Planck's function, σ the Stefan-Boltzmann constant, and $E_3(x)$ the exponential integral. The optically thin limit is applicable for conditions of the present study, from which it follows that ε is proportional to τ_{vis} . Employing eqs. (6) and (7), together with our evaluation of β_{IR} , we find by numerical integration that

$$\varepsilon = 0.0816 \tau_{\text{vis}} \quad (8)$$

for $T = 217 \text{ K}$.

The corresponding slab absorptivity is

$$\alpha = \frac{1}{\sigma T_e^4} \int_0^\infty F_\lambda [1 - 2E_3(\tau_{\text{IR}})] d\lambda \quad (9)$$

where F_λ is the spectral infrared flux incident upon the bottom of the layer and T_e the effective temperature of the planet. The above assumes that emission by the atmosphere overlying the aerosol layer is negligible, as will be later justified. While it is possible to calculate F_λ from known atmospheric opacities in the absence of clouds, the presence of clouds in the real atmosphere renders this approach currently impractical. Cirrus clouds, for example will exert a significant influence upon F_λ , but radiative properties of cirrus clouds are not well known. As an alternative we will simply let $F_\lambda = e_\lambda(T_e)$ in this present first-order analysis, with $T_e = 253 \text{ K}$ denoting the effective temperature of the planet, and eqs. (6) and (9) yield

$$\alpha = 0.1029 \tau_{\text{vis}} \quad (10)$$

Note that $\alpha/\varepsilon = 1.26$, the departure of this ratio from unity being a measure of the non-grayness of the aerosol layer.

Stratospheric temperature at 20 km

To estimate the stratospheric temperature in the vicinity of the 20 km aerosol layer, we neglect any influence due to dynamics. If important, this should contribute to heating on a global average, since the divergence of the total vertical eddy flux would appear to be negative at this altitude (Palmén & Newton, p. 59, 1969). The stratospheric temperature is consequently assumed to be governed by radiative transfer due both to gaseous and aerosol opacities. For these separate constituents we shall employ the Newtonian cooling expression

$$Q = \varrho c_p N (T - T_0) \quad (11)$$

where Q represents a perturbed radiative cooling rate per unit volume relative to an equilibrium state at temperature T_0 , N is the Newtonian cooling coefficient, while ϱ and c_p are the atmospheric density and specific heat, respectively.

1. Newtonian cooling coefficients

Consider first the evaluation of N for the gaseous constituents, with H_2O and CO_2 comprising the main infrared opacity sources at this altitude. Letting q_R represent the net infrared flux, measured in the positive z -direction, the infrared cooling per unit volume is

$$\frac{dq_R}{dz} = -\sigma T^4 \frac{d\varepsilon_g}{dz} \quad (12)$$

where ε_g is the gas emissivity and z is altitude. This formulation is simply the direct cooling to space approximation first suggested by Rodgers & Walshaw (1966), the absence of exchange integrals being due to the near isothermality of the stratosphere. Further discussion as to the neglect of exchange integrals is given by Cess & Ramanathan (1972), Cess & Harshvardhan (1974), and Ramanathan (1974).

If we perturb eq. (12) about an equilibrium state by employing $T^4 - T_0^4 = 4T_0^3(T - T_0)$, comparison of the perturbed volumetric cooling with eq. (11) yields

$$N = -\frac{4\sigma T_0^3}{\varrho c_p} \frac{d\varepsilon_g}{dz} \quad (13)$$

A suitable expression for the water vapor emissivity applicable to the stratosphere is given by Cess (1974) as

$$\varepsilon_g = 0.072 \sqrt{r P_w H P / 2}$$

where H is the atmospheric scale height (cm), P_w the partial pressure (atm) of H_2O , P the total pressure (atm) with $P/2$ constituting the Curtis-Godson broadening pressure, and we take the diffusivity factor to be $r=1.8$, appropriate for strong rotational lines (Grief & Habib, 1969; Hottel & Sarofim, p. 285, 1967).

Assuming a hydrostatic atmosphere for the evaluation of $d\varepsilon_g/dz$, it follows that

$$N(H_2O) = \frac{0.204\sigma T^3}{\rho c_p H} \sqrt{r P_w P H}$$

With $P = 0.055$ atm, $T = 217$ K and $H = 6.35 \times 10^5$ cm at the 20 km level, together with $P_w/P = 4.8 \times 10^{-4}$, then $N(H_2O) = 0.0022$ day $^{-1}$.

For CO_2 the emissivity may be expressed as

$$\varepsilon = \frac{e_{\lambda_0}(T)}{\sigma T^4} A$$

where λ_0 is the wavelength of the 15 μ m band while A is the band absorptance, given by Cess & Ramanathan (1972) for conditions appropriate to the stratosphere as

$$A = 2A_0 \ln(1 + \sqrt{\xi})$$

with $\xi = 2rSH\gamma_0 P_c P / A_0 d$, where P_c is the partial pressure of CO_2 , the band intensity is taken as $S = 242$ (300/ T) cm $^{-2}$ atm $^{-1}$, and $A_0 = 22.3$ ($T/300$) $^{1/2}$ cm $^{-1}$, $\gamma_0 = 0.097$ (300/ T) $^{1/2}$ cm $^{-1}$ atm $^{-1}$, and $d = 1.56$ cm $^{-1}$ (Cess & Ramanathan, 1972). Again $r=1.8$ is appropriate. Following the same procedure as before, except that here we expand $e_{\lambda_0}(T)$ about $e_{\lambda_0}(T_0)$, then

$$N(CO_2) = \frac{2A_0}{\rho c_p H} \frac{de_{\lambda_0}(T)}{dT} \left(\frac{\xi}{\sqrt{\xi} + \xi} \right) \quad (14)$$

For $\xi < 1$ (nonoverlapping lines), this reduces to the same expression as that of Cess & Harshvardhan (1974). With $P_c/P = 0.0003$ and for conditions at the 20 km level, $N(CO_2) = 0.0096$ day $^{-1}$.

The value of N for both gaseous constituents is simply the sum of the separate quantities,

Tellus XXVIII (1976), 1

band overlap being negligible at this altitude, such that

$$N(\text{gas}) = 0.012 \text{ day}^{-1} \quad (15)$$

We note that the preceding constitutes a more direct determination of the Newtonian cooling coefficients than that presented, for example, by Goody (1964) or Goody & Belton (1967). In addition, eq. (15) is in excellent agreement with an estimate for N based upon the local cooling rate at 20 km. Denoting the cooling rate by dT/dt , and noting from eq. (12) that

$$\rho c_p \frac{dT}{dt} = -\sigma T^4 \frac{d\varepsilon_g}{dz}$$

then employing eq. (13)

$$N = \frac{4}{T} \frac{dT}{dt}$$

From Ramanathan (1974), Manabe & Strickler (1964), or Dopplick (as reported in *Man's impact on the global environment*, 1971, MIT Press, p. 86), $dT/dt = 0.7$ K/day at 20 km, such that with $T = 217$ K, then $N = 0.013$ day $^{-1}$ which is consistent with eq. (15). We have not included the 9.6 μ m ozone band in our formulation for N , since the contribution of this band is to heating through absorption of upward infrared radiation, and thus the 9.6 μ m O_3 band will influence the equilibrium temperature T_0 but not N . This conclusion is consistent with the results of Dickinson (1973).

Turning next to the aerosol layer, the volumetric cooling of this layer may be expressed as

$$\frac{dq_R}{dz} = \frac{2\varepsilon\sigma T^4 - \alpha\sigma T_e^4}{L}$$

where $L = 2.3$ km is the aerosol scale dimension as previously discussed. Proceeding as before, it is easily found that

$$N(\text{aerosol}) = \frac{8\varepsilon\sigma T^3}{\rho c_p L}$$

or employing eq. (8)

$$N(\text{aerosol}) = 0.159 \tau_{vis} \text{ day}^{-1} \quad (16)$$

We caution, however that this result is very approximate, since L depends upon the aerosol mass density, and this is not well known.

2. Energy balance at 20 km

A radiative energy balance at the 20 km level yields

$$N(\text{gas})[T_A - T_0(\text{gas})] + N(\text{aerosol})[T_A - T_0(\text{aerosol})] = 0 \quad (17)$$

where T_A is the 20 km temperature with both gas and aerosol opacities taken into account. The equilibrium temperature for the gas simply corresponds to the 20 km temperature in the absence of aerosol opacity, and we take this to be $T_0(\text{gas}) = 214$ K (Ramanathan, 1974). Conversely, a radiative equilibrium balance for the aerosol layer yields

$$(T_0/T_e)^4 = \alpha/2\varepsilon$$

such that with $T_e = 253$ K and $\alpha/\varepsilon = 1.26$, then $T_0(\text{aerosol}) = 225$ K. It now follows from eq. (15), (16) and (17) that

$$T_A = 214 \left(\frac{1 + 13.90 \tau_{\text{vis}}}{1 + 13.22 \tau_{\text{vis}}} \right) \quad (18)$$

For present conditions ($\tau_{\text{vis}} = 0.02$), $T_A = 216.3$ K, indicating that aerosols contribute about 2 K of heating at the 20 km level. Since $T_A \leq 225$ K, the maximum additional increase that could be achieved through a large increase in aerosol concentration would be roughly 9 K, a result which has interesting implications, as discussed in the last section of the paper, concerning events following the eruption of Mount Agung. It should be noted that the increase in stratospheric temperature due to the aerosol layer is a consequence of the layer being nongray ($\alpha > \varepsilon$). If we had instead assumed a gray aerosol ($\alpha = \varepsilon$), then $T_0(\text{aerosol}) = 213$ K, essentially the same as $T_0(\text{gas})$, and the aerosol would not alter the 20 km temperature. The preceding neglects changes in T_e as a consequence of the aerosol reflectivity, but this will be a small effect and will not influence our qualitative conclusions.

Global energy balance

We now apply the results of the preceding section to a global energy balance, in order to obtain a first-order estimate as to the magnitude of the reduction in global mean surface

temperature as a function of aerosol optical thickness, τ_{vis} .

Solar irradiation of the aerosol layer consists of direct radiation from above and reflected radiation from below. We denote the albedo of the earth-atmosphere system in the absence of the aerosol layer by α , and we may assume that the contribution to this quantity from the atmosphere overlying the aerosol layer is negligible, since the Rayleigh scattering albedo for the atmosphere above 20 km is of order 10^{-3} . We further let I_s denote the solar constant ($1.95 \text{ cal cm}^{-2} \text{ min}^{-1}$) while S is the solar flux absorbed by the earth-atmosphere system, and recall that R is the reflectivity of the aerosol layer. Accounting for multiple reflections between the earth-atmosphere system and the aerosol, then

$$S = (I_s/4)(1 - \alpha)(1 - R)[1 + (aR) + (aR)^2 + \dots]$$

or

$$S = S_0 \left(\frac{1 - R}{1 - aR} \right) \quad (19)$$

where $S_0 = (I_s/4)(1 - \alpha)$ is the absorbed solar flux in the absence of the aerosol.

With F denoting the upward infrared flux incident upon the bottom of the aerosol layer, a global energy balance yields

$$S_0[(1 - R)/(1 - aR)] = F(1 - \alpha) + \varepsilon \sigma T_A^4 \quad (20)$$

Inherent in this expression is the assumption that the atmosphere overlying the aerosol layer produces a negligible contribution to the infrared energy budget of the earth-atmosphere system. This is consistent with model atmosphere calculations, from which we estimate that the contribution is roughly 2%.

Eq. (20) describes the outgoing flux F in terms of known quantities together with aerosol parameters which are expressed as a function of τ_{vis} . It remains to relate F to surface temperature, and for this purpose we let

$$F = F_0 + \frac{dF}{dT_s} \Delta T_s \quad (21)$$

where F_0 is the outgoing infrared flux in the absence of the aerosol layer, while ΔT_s is the change in surface temperature as a consequence of the aerosol. Since $F_0 = S_0$, it follows from

eqs. (19), (20) and (21) that

$$\Delta T_s = -\frac{S_0}{dF/dT_s} \times \left\{ 1 - \frac{[(1-R)/(1-aR)] - \epsilon \sigma T_A^4/S_0}{1-\alpha} \right\} \quad (22)$$

We shall employ Budyko's (1969) empirical expression to evaluate dF/dT_s , which gives (with F in $\text{cal cm}^{-2} \text{ min}^{-1}$)

$$\frac{dF}{dT_s} = 0.0032 - 0.0023 A_c \quad (23)$$

where A_c is the fraction of cloud cover, and for average cloudiness we take $A_c = 0.5$. This does not account for cloud cover as a feedback mechanism, a point which is discussed in the following section.

Budyko's empirical flux formulation is based upon monthly averages of 260 meteorological stations, and this type of averaging has been criticized as not correctly describing F as a function of T_s on a global mean basis. In this regard we note that model atmosphere calculations for $A_c = 0$ are consistent with eq. (23), since these yield $dF/dT_s = 0.0033$ (Manabe & Wetherald, 1967) and 0.0028 (Cess, 1974). This gives us some confidence in the applicability of eq. (23).

A further point is that eq. (23) does not account for perturbations in stratospheric temperature as a result of increased aerosol concentration. To assess the possible importance of this effect, we have used the model calculations of Cess (1974, 1975), employing a single effective fixed-temperature cloud; a tropospheric lapse rate of 6.5 K/km was chosen together with an isothermal stratosphere. The model calculations indicate that dF/dT_s changes by only 6% for an increase in stratospheric temperature from 216 K to 225 K, from which we conclude that aerosol induced stratospheric temperature perturbations will not significantly influence dF/dT_s .

The surface temperature change, described by eq. (22), is illustrated in Fig. 1 as a function of τ_{vis} . Also illustrated in this figure is the change in surface temperature if the infrared opacity (greenhouse effect) of the aerosol layer is neglected ($\alpha = \epsilon = 0$). The comparison clearly illustrates that the aerosol infrared opacity

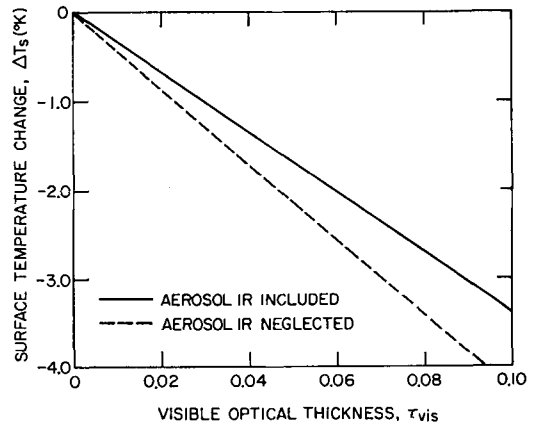


Fig. 1. Reduction in global mean surface temperature as a function of the visible optical thickness of stratospheric aerosols.

plays a small role in compensating for the reduction in surface temperature due to the aerosol solar reflectivity. The results further indicate that the normal stratospheric aerosol ($\tau_{\text{vis}} = 0.02$) reduces the global mean surface temperature by 0.7 K; a doubling of aerosol concentration would produce a further decrease in surface temperature by the same amount. As previously discussed, there is considerable uncertainty in our calculation of T_A , which appears in eq. (22), but errors in T_A produce an insignificant effect upon the ΔT_s results.

Discussion of results

The present analysis predicts that increases in stratospheric aerosol concentration will produce an increase in atmospheric temperature in the vicinity of the aerosol layer, the model calculations indicating that this increase could be as large as 9 K, but the important point is that a qualitatively significant increase in temperature should occur. This is consistent with events following volcanic activity. Newell (1970) has analyzed radiosonde temperature observations from Australian stations following the eruption of Mount Agung (latitude 8°S) in 1963. The temperature at the 19.5 km level increased approximately 6 K, maintained this level for a year, and remained 2 to 3 K above the pre-Agung level for several years. This temperature enhancement was less pronounced

at lower altitudes, rarely exceeding 2 K at the 12.4 km level.

Many observations have been performed which are indicative of changes in the visible optical thickness of the aerosol layer following volcanic activity, as summarized by Volz (1970). Again choosing $\tau_{\text{vis}} = 0.02$ for the normal stratosphere, these indicate that for about a year following the Agung eruption τ_{vis} was increased by a factor of roughly two (47° N) to three (19° N) within the Northern Hemisphere. This latitude dependence is consistent with Rosen's (1968) conclusion that aerosol concentration decreases with increasing latitude. Pueschel et al. (1972), measuring normal incident radiation at Mauna Loa Observatory (19° N), found that reduced atmospheric transmissivities persisted for roughly seven years after the Agung eruption, in agreement with Castleman (1974), who has shown that the lesser eruptions of Mount Taal (1965, 14° N) and Mount Awu (1966, 3° N) contributed significantly to the sulfate inventory of the Northern Hemisphere stratosphere. A considerably greater increase in τ_{vis} occurred in the Southern Hemisphere (Volz, 1970), with τ_{vis} increasing by an average of roughly a factor of eight (25° to 38° S) for the one-year period following Agung. Once again this is consistent with sulfate concentration measurements (Castleman, 1974; Castleman et al., 1974), which show considerably greater stratospheric sulfate concentrations for the Southern as compared to the Northern Hemisphere for the post-Agung period.

These observed increases in τ_{vis} will of course not give rise to a cooling of the planet of the magnitude indicated in Fig. 1, since the ΔT_s results refer to a steady state. The response of the planet to aerosol-induced changes in the global energy balance would require a sustained aerosol enhancement over a long period of time in order to achieve steady-state conditions. Volcanic activity during the 1960's could, however, contribute in part to what appears to be a global cooling trend since 1940 (e.g. Budyko, 1969).

The response of the stratosphere, on the other hand, is quite rapid. The 6 K increase in temperature at the 20 km level, observed by Newell (1970) following the Agung eruption, corresponds approximately to the same latitude range as the observed eight-fold average increase in τ_{vis} . This temperature increase was

achieved in roughly three months. It is interesting to note that if we increase τ_{vis} from 0.02 to 0.16, eq. (18) yields a 5 K increase in temperature at the 20 km level. We caution again, however, that our stratospheric temperature estimates are crude, so that this apparent agreement could be accidental.

As a final point we wish to emphasize that our first-order global energy balance, the results of which are illustrated in Fig. 1, neglects what might be a potentially important atmospheric feedback mechanism. We have arbitrarily assumed that the fraction of cloud cover is constant ($A_c = 0.5$) and thus independent of surface temperature, but this is probably not the case. For example if cloud amount were to increase with decreasing global surface temperature, this would produce a positive feedback mechanism (Schneider, 1972), and the reduction in surface temperature as a function of τ_{vis} would be more severe than that depicted in Fig. 1.

Although Paltridge (1974) points out that there is an observed decrease in global cloud amount of about five percent between the times of minima and maxima in the annual cycle of sun-earth distance, this does not necessarily imply that global cloud amount decreases with decreasing global surface temperature (negative feedback), since the phase lag in sea-surface temperature is nearly three months (Palmén & Newton, p. 475, 1969). We feel that cloud amount as a feedback mechanism is as yet unresolved.

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Note added in proof

Our eq. (5) for aerosol reflectivity refers to a single-wavelength calculation at $\lambda = 0.55 \mu\text{m}$.

More recently we have calculated the solar reflectivity for the range of solar wavelengths, and upon averaging this over the solar spectrum we obtain $R = 0.30 \tau_{\text{vis}}$. Comparison with eq. (5)

illustrates that the $0.55 \mu\text{m}$ expression does not differ substantially from the solar-averaged result.

REFERENCES

- Budyko, M. I. 1969. The effect of solar radiation variations on the climate of the Earth. *Tellus* 21, 611-619.
- Castleman, A. W., Jr. 1974. Nucleation processes and aerosol chemistry. *Space Science Reviews* 15, 547-589.
- Castleman, A. W., Jr, Munkelwitz, H. R. & Manowitz, B. 1974. Isotopic studies of the sulphur component of the stratospheric aerosol layer. *Tellus* 26, 222-234.
- Cess, R. D. 1974. Radiative transfer due to atmospheric water vapor: Global considerations of the earth's energy balance. *J. Quant. Spectrosc. Radiat. Transfer* 14, 861-871.
- Cess, R. D. 1975. Global climate change: an investigation of atmospheric feedback mechanisms. *Tellus* 27, 193-198.
- Cess, R. D. & Harshvardhan. 1974. Shear-flow stability within the atmosphere of Venus. *J. Fluid Mech.* 66, 267-272.
- Cess, R. D. & Ramanathan, V. 1972. Radiative transfer in the atmosphere of Mars and that of Venus above the cloud deck. *J. Quant. Spectrosc. Radiat. Transfer* 12, 933-945.
- Coakley, J. A., Jr & Chylek, P. 1975. The two-stream approximation in radiative transfer: including the angle of the incident radiation. *J. Atmos. Sci.* 32, 409-418.
- Coakley, J. A., Jr & Grams, G. W. 1975. Relative influence of visible and infrared optical properties of a stratospheric aerosol layer on the global climate. Preprint.
- Deirmendjian, D. 1969. *Electromagnetic Scattering on spherical polydispersions*. American Elsevier Publishing Co., New York, 290 pp.
- Dickinson, R. E. 1973. Method of parameterization for infrared cooling between altitudes of 30 and 70 kilometers. *J. Geophys. Res.* 78, 4451-4457.
- Elterman, L., Toolin, R. B. & Essex, J. D. 1973. Stratospheric aerosol measurements with implications for global climate. *Appl. Optics* 12, 330-337.
- Goody, R. M. 1964. *Atmospheric radiation*. Oxford University Press, London, 436 pp.
- Goody, R. M. & Belton, M. J. S. 1967. Radiative relaxation times for Mars (a discussion of Martian atmospheric dynamics). *Planet. Space Sci.* 15, 247-256.
- Grief, R. & Habib, I. S. 1969. Infrared radiation transport: exact and approximate results. *J. Heat Transfer* 91, 282-284.
- Hottel, H. C. & Sarofim, A. F. 1967. *Radiative transfer*. McGraw-Hill, New York, 520 pp.
- Lazrus, A. L., Gandrud, B. & Cadle, R. D. 1971. Chemical composition of air filtration samples of the stratospheric sulfate layer. *J. Geophys. Res.* 76, 8083-8088.
- Manabe, S. & Strickler, R. F. 1964. Thermal equilibrium of the atmosphere with a convective adjustment. *J. Atmos. Sci.* 21, 361-385.
- Manabe, S. & Wetherald, R. T. 1967. Thermal equilibrium of the atmosphere with a given distribution of relative humidity. *J. Atmos. Sci.* 24, 241-259.
- Newell, R. E. 1970. Stratospheric temperature change from the Mt. Agung volcanic eruption of 1963. *J. Atmos.* 27, 977-978.
- Palmen, E. & Newton, C. W. 1969. *Atmospheric circulation systems*. Academic Press, New York, 603 pp.
- Palmer, K. F. & Williams, D. 1975. Optical constants of sulfuric acid; application to the clouds of Venus? *Appl. Optics* 14, 208-219.
- Paltridge, C. W. 1974. Global cloud cover and earth surface temperature. *J. Atmos. Sci.* 31, 1571-1576.
- Penndorf, R. B. 1962. Scattering and extinction coefficients for small absorbing and nonabsorbing aerosols. *J. Opt. Soc. Am.* 52, 896-904.
- Plass, G. N. 1966. Mie scattering and absorption cross sections for absorbing particles. *Appl. Optics* 5, 279-285.
- Pueschel, R. F., Machta, L., Cotton, G. F. & Flowers, E. C. 1972. Normal incidence radiation trends on Mauna Loa, Hawaii. *Nature* 240, 545-547.
- Ramanathan, V. 1974. A simplified stratospheric radiative transfer model: theoretical estimates of the thermal structure of the basic and perturbed stratosphere. *Preprints of the Second International Conference on the Environmental Impact of Aerospace Operations in the High Stratosphere*. American Meteorological Society.
- Rasool, S. I. & Schneider, S. H. 1971. Atmospheric carbon dioxide and aerosols: effect of large increases on global climate. *Science* 173, 138-141.
- Reck, R. A. 1974. Influence of surface albedo on the change in the atmospheric radiation balance due to aerosols. *Atmospheric Environment* 8, 823-833.
- Remsberg, E. E. 1973. Stratospheric aerosol properties and their effects on infrared radiation. *J. Geophys. Res.* 78, 1401-1408.
- Rodgers, C. D. & Walshaw, C. D. 1966. The computation of infrared cooling rate in planetary atmospheres. *Quart. J. Roy. Met. Soc.* 92, 67-92.
- Rosen, J. M. 1968. Simultaneous dust and ozone soundings over North and Central America. *J. Geophys. Res.* 73, 479-486.
- Rosen, J. M. 1971. The boiling point of stratospheric aerosols. *J. Appl. Meteor.* 10, 1044-1046.
- Schneider, S. H. 1972. Cloudiness as a global climatic feedback mechanism: the effects on the radiation balance and surface temperature on variations in cloudiness. *J. Atmos. Sci.* 29, 1413-1422.

- Toon, O. B. & Pollack, J. B. 1973. Physical properties of the stratospheric aerosols. *J. Geophys. Res.* 78, 7051–7056.
- van de Hulst, H. C. 1957. *Light scattering by small particles*. John Wiley, New York, 470 pp.
- Volz, F. E. 1970. Atmospheric turbidity after the Agung eruption of 1963 and size distribution of the volcanic aerosol. *J. Geophys. Res.* 75, 5185–5193.
- Wang, W. C. & Domoto, G. A. 1974. The radiative effect of aerosols in the earth's atmosphere. *J. Appl. Meteor.* 13, 521–534.
- Yamamoto, G. & Tanaka, M. 1972. Increase of global albedo due to air pollution. *J. Atmos. Sci.* 29, 1405–1412.

СТРАТОСФЕРНЫЕ АЭРОЗОЛИ: ВЛИЯНИЕ НА ТЕМПЕРАТУРУ АТМОСФЕРЫ И ГЛОБАЛЬНЫЙ КЛИМАТ

Распространенным является мнение, что значительная часть аэрозольной популяции стратосферы состоит из сульфатов, предположительно, в форме серной кислоты. Увеличение аэрозоля в стратосфере может влиять на климат путем изменения величины отражения солнечного света, так же как и усилением парникового эффекта атмосферы — двумя конкурирующими механизмами в изменении глобальной температуры поверхности. Предполагая, что аэрозоль состоит из переохлажденного водного 75 % раствора серной кислоты, мы даем оценку в первом порядке влияния концентрации аэрозоля на глобальную температуру поверхности. Модельные расчеты показывают, что увеличение отраже-

ния солнечного света представляет доминирующий вклад аэрозоля. Нормальные концентрации аэрозоля уменьшают глобальную температуру поверхности примерно на 0,7 К, а удвоение концентрации приведет к дальнейшему падению температуры на ту же величину. Увеличение концентрации аэрозоля приводит к нагреву стратосферы путем поглощения инфракрасной радиации, излучаемой системой земля–атмосфера. Модель показывает, что температура стратосферы на уровне 20 км может быть повышена на величину до 9 К, что согласуется с наблюдениями в Южном полушарии, последовавшими за извержением вулкана Агунг.