Influence of the Atmospheric Aerosol on Albedo

DAVID S. ENSOR, WILLIAM M. PORCH, MICHAEL J. PILAT AND ROBERT J. CHARLSON

Depts. of Civil Engineering and Geophysics, University of Washington, Seattle

ABSTRACT

The possible climatic effects of the secular increase of aerosols from man's activities have been coupled with the microphysics of the aerosol properties. The magnitude of the critical aerosol absorption coefficient to backscatter coefficient, $(b_{abs}/b_{bs})_{critical}$, was estimated for a model atmosphere corresponding to cooling or heating of the earth with increasing aerosol concentration. The b_{abs}/b_{bs} ratio was calculated with Mie theory assuming a Junge particle size distribution and spherical particles as a function of the imaginary part of the particle refractive index (particle light absorption) and the size distribution slope. Comparing the b_{abs}/b_{bs} ratio calculated from Mie theory to the critical b_{abs}/b_{bs}, cooling might ensue if the imaginary part is less than 10^{-3} while heating may result if it is greater than 0.1.

1. Introduction

The effect of the increasing atmospheric aerosol concentration (due to increased anthropogenic production of particulate air pollutants) upon the albedo has been questioned for some time. McCormick and Ludwig (1967) suggested that an increasing concentration of atmospheric aerosols could increase the albedo and thus cool the earth. Bryson (1968) also discussed this possibility. Charlson and Pilat (1969) reported that an increase in the atmospheric aerosol concentration could either cause cooling or heating of the earth-atmosphere system, depending on the relative magnitudes of the aerosol absorption and backscattering coefficients. The general aspects of climate change have been discussed by Landsberg (1970) and in the Report of the Study of Critical Environmental Problems (1970). This paper presents calculated light absorption and backscattering coefficients for model atmospheric aerosols and discusses the apparent effects of aerosols on albedo.

2. Development of earth-space heat transfer equations

The atmospheric aerosol can affect the transfer of radiant energy between the earth-atmosphere system and outer space by 1) reflecting (scattering) incoming radiation, 2) reflecting (scattering) outgoing radiation, 3) absorbing incoming radiation, and 4) absorbing outgoing radiation. Let us consider the effect of the atmospheric aerosol upon the amount of energy absorbed by the earth-atmosphere system. The amount of energy absorbed per unit time (W) by the earth-atmosphere system due to aerosols is the sum of the absorption due to aerosol-diminished radiation by the earth's surface and by the atmosphere, i.e.,

$$W_{earth-atmos} = W_{surface} + W_{atmos}.$$

Expressing each of these terms as a function of atmospheric and surface properties, the first can be expressed as

$$W_{surface} = S(1-A),$$

where A is the albedo of the surface (radiation reflected/radiation incident), and S the solar energy per unit time at the surface of the earth; and if we neglect absorption of surface reflected radiation with a flat earth approximation, the second may be written as

$$W_{atmos} = S_0[1 - \exp(-b_{abs}X)],$$

where b_{abs} is the absorption coefficient of the aerosol, X the light path length through the atmosphere, and S_0 the energy per unit time incident on the atmosphere. Longwave terrestrial radiation has been neglected in this simple model. Substituting for $W_{surface}$ and W_{atmos} in (1), we have

$$
\begin{aligned}
W_{earth-atmos} = & (1-A)S_0 \exp[-(b_{bs}+b_{abs})X] \\
& \text{(surface heating)} \\
& + S_0[1 - \exp(-b_{abs}X)], \quad (4) \\
& \text{(atmospheric heating)}
\end{aligned}
$$

where b_{bs} is the backscattering coefficient of the aerosol, undefined as to specific angles. Assuming that the fractional radiation absorption at the surface, $(1-A)$, remains constant with varying atmospheric aerosol concentration, Eq. (4) can be used to estimate whether an increased particulate concentration will heat or cool the earth-atmosphere system.

The atmosphere can usually be considered to be optically thin for aerosol absorption and backscatter. Where $b_{abs}X$ and $b_{bs}X$ are much less than 1, the exponential terms can be approximated by

$$\exp(-b_{abs}X) \approx 1 - b_{abs}X \quad \text{and} \quad \exp(-b_{bs}X) \approx 1 - b_{bs}X.$$

Eq. (4) thus reduces to

$$W_{\text{earth-atmos}} \approx (1-A)S_0(1 - b_{bs}X - b_{abs}X) + S_0 b_{abs}X. \quad (5)$$

The difference in energy between the earth-atmosphere system with and without aerosols is therefore

$$\Delta W_{\text{earth-atmos}} = -(1-A)S_0(b_{bs} + b_{abs})X + S_0 b_{abs}X. \quad (6)$$

A criteria which may be used to determine if the secular increase in aerosol heats or cools the earth-atmosphere system can be obtained by solving (6) for b_{abs}/b_{bs}. The ratio of absorption to backscatter is given by

$$\frac{b_{abs}}{b_{bs}} \begin{matrix} > \\ = \\ < \end{matrix} \frac{(1-A)}{A}, \quad (7)$$

where "greater than" implies increasing energy or heating of the earth with increasing aerosol, "equals" implies no change in energy with increasing aerosol, and "less than" implies decreasing energy or cooling of the earth with increasing aerosol. Thus, from a simple model considering only gross radiative transfer effects, the critical aerosol absorption to backscatter ratio (b_{abs}/b_{bs}), which may be used as a criteria for heating or cooling of the earth, is a function only of the albedo of the surface. However, (7) may be too strict a requirement for the critical effective absorption-to-backscatter ratio because the effects of absorption and backscatter of primary surface reflected radiation may be important. Also, in areas where surface moisture is a factor not all the energy is available for sensible heating. Mitchell (1971) has calculated the critical aerosol absorption-to-backscatter ratio after separating sensible from latent heating. He also includes backscatter of primary surface reflected radiation and the fact that the temperature of the atmosphere in close vicinity to the surface of the earth is due only to aerosol introduced into the air in convectively active communication with the surface. The critical ratio with the same criteria for heating and cooling as before becomes

$$\frac{b_{abs}}{b_{bs}} \begin{matrix} > \\ = \\ < \end{matrix} \frac{C(1-A)(1-AK)}{D(1+A) - C(1-A)}, \quad (8)$$

where K is the ratio of effective backscattering coefficient for surface reflected radiation to b_{bs}, C the sensible heating of the surface of the earth as a fraction of the total solar heating of the surface, and D the fraction of the aerosol in convective contact with the surface of the earth. The values of K and D are quite difficult to determine accurately and at present can only be estimated. For areas with small A and C, and where D and K are both of order unity, Eqs. (8) and (7) reduce to the two equations derived for a thin aerosol layer by Atwater (1970). Mitchell proposed a tentative average value of $D = 0.75$ for the total tropospheric aerosol. For a homogeneous aerosol layer the value of K varies from $K = 0$ (where absorption dominates backscatter of surface reflected radiation) to $K \approx 1$ (where back scatter dominates absorption of surface reflected radiation). In fact, the effective backscattering coefficient for upward directed radiation may exceed that for downward directed radiation in cases where the aerosol characteristics are distributed sufficiently heterogeneously with height.

Table 1 shows values of the critical ratio calculated with the use of Eq. (7) and those reported by Mitchell from Eq. (8). The values for K and D were assumed to be 1 and 0.75, respectively. From Table 1, the simple model considering only radiative transfer is much more stringent than Mitchell's more sophisticated one. A much smaller ratio is sufficient to produce atmospheric heating in Mitchell's model. For the earth surfaces where moisture is important, the critical ratio is less than 0.5 (0.1 for oceans), while for dryer surfaces this ratio is of order unity.

With these simple heat balance models, the order of magnitude of the critical (b_{abs}/b_{bs}) ratio can be estimated. Very little data exists from which the absorption-to-backscatter ratio can be estimated for the background aerosol. The limited measurements of background aerosol light scattering have been summarized by Porch et al. (1970) for a few remote locations. In the absence of experimental data, a calculation of the critical ratio for aerosols of various realistic size distributions and refractive indices should provide insight into the possible effects of increasing aerosol concentrations on climate.

TABLE 1. Comparison of critical absorption-to-backscatter ratios for different surface types as derived in this paper using $(1-A)/A$ [Eq. (7)] and by Mitchell [Eq. (8)].

Surface type	Albedo A	Sensible heating index C	Critical ratio b_{abs}/b_{bs}	
			$(1-A)/A$	Mitchell's model
Urban areas	0.20	0.80	4	1.97
Deserts	0.30	0.40	2.3	1.50
Prairies and grasslands (warm season)	0.20	0.40	4	0.44
Forests	0.16	0.30	5.2	0.34
Oceans (mid-latitude)	0.08	0.09	12	0.10
Snowfields (stable)	0.70	0.09	0.43	<0.01

3. Calculation of b_{abs} and b_{bs}

a. Theory

The ratio of the absorption coefficient to the backscatter coefficient was calculated assuming an atmospheric aerosol size distribution model. A power law atmospheric aerosol size distribution (Junge, 1963) was used to describe the number frequency distribution $n(r)$ for particle radii ranging from 0.01 to 10 μ, i.e.,

$$n(r) = \frac{dn}{dr} = Cr^{-(\beta+1)}, \qquad (9)$$

using exponents β of 2, 3 and 4. These values of the exponent are typical in the atmosphere (Charlson, 1969).

The backscatter coefficient for spherical particles is related to the backscatter efficiency factor Q_{bs} by

$$b_{bs} = \int_{r_1}^{r_2} Q_{bs} \pi r^2 n(r) dr. \qquad (10)$$

The backscatter efficiency factor is defined as

$$Q_{bs} = \frac{1}{\alpha^2} \int_{\pi/2}^{\pi} [i_1(\theta) + i_2(\theta)] \sin\theta d\theta, \qquad (11)$$

where α is the size parameter $(2\pi r/\lambda)$, r the particle size, λ the wavelength of light, θ the angle of scattered light with respect to the incident beam, and $i_1(\theta)$, $i_2(\theta)$ the intensity of scattered light per solid angle polarized perpendicular or parallel, respectively, to the plane of scattering. The absorption coefficient is related to the absorption efficiency factor Q_{abs} by

$$b_{abs} = \int_{r_1}^{r_2} Q_{abs} \pi r^2 n(r) dr, \qquad (12)$$

where the absorption efficiency factor is defined in terms of the extinction efficiency factor Q_{ext} and the scattering efficiency factory by the relation

$$Q_{abs} = Q_{ext} - Q_{scat}. \qquad (13)$$

The various efficiency factors are given by

$$Q_{ext} = \frac{2}{\alpha^2} \sum_{t=1}^{n} (2t+1) \, \mathrm{Re}(a_t + b_t), \qquad (14)$$

$$Q_{scat} = \frac{2}{\alpha^2} \sum_{t=1}^{n} (2t+1)(|a_t|^2 + |b_t|^2), \qquad (15)$$

where Re means the real part of the quantity in the parenthesis and a_t and b_t are complex Mie amplitude coefficients. These equations can be calculated for Mie theory as summarized by van de Hulst (1957).

Eqs. (10)–(12) were numerically integrated with the Romberg integration technique described by Wilf (1967).

b. Aerosol refractive index

The important parameter is the refractive index of the aerosol. The refractive index, $m = n_1 - n_2 i$, consists of the real part n_1 describing scattering and the imaginary part n_2 describing the light absorption of the particles. The real part seems to be relatively well defined at about 1.5 ± 0.2 (Bullrich, 1964). However, the imaginary part, crucial for the heat balance, is not well known. In principle, the imaginary part of the refractive index could vary from 0.0 for materials such as sea salt to about 0.7 for opaque materials such as soot or metal oxide dust. Eiden (1966, 1971) reported an indirect determination of the imaginary refractive index. Angular polarization measurements of a volume of atmospheric aerosol were fitted using Mie theory and assumed size distribution and refractive indices. The best fit for dry aerosol was $m = 1.50 - (0.01 < n_2 < 0.1)i$ and for high humidity conditions both n_1 and n_2 decreased in magnitude. Fischer (1970) reported that the magnitude of the imaginary refractive index is 10^{-2} from measurements of the light scattered from aerosol particle deposits.

At this time, no technique exists to directly measure *in situ* the absorption of a volume of atmospheric aerosol.

4. Discussion of results

The absorption coefficient to backscatter coefficient ratios calculated for various exponents of the size distribution and the imaginary part of the refractive index are summarized in Table 2. The imaginary part of the refractive index is of primary importance while the size distribution parameter is secondary.

The critical (b_{abs}/b_{bs}) ratios in Table 1 can be compared to the calculated ratios in Table 2. Very qualitatively, we can say that if the worldwide absorption coefficient is described by $n_2 < 10^{-3}$, the effect of a secular increase in aerosol would be to cool the earth; if $10^{-3} < n_2 < 10^{-1}$, the effect would be ambiguous, and if $n_2 > 10^{-1}$ the effect would be to heat the earth, regardless of choice of climate model. If Mitchell's (1971) model is chosen over the simple model herein, the range of

TABLE 2. Ratio of the absorption coefficient to the backscattering coefficient (b_{abs}/b_{bs}) for a wavelength of 500 nm.*

m	β 2.0	3.0	4.0	Effect†
n_1 n_2				Model in this paper Mitchell's model
1.50–0i	0.0	0.0	0.0	——Cooling——
1.5–10⁻⁴i	0.074	0.014	0.017	
1.5–10⁻³i	0.62	0.14	0.17	/////////// /////////// ambiguous
1.5–0.01i	3.9	1.2	1.7	ambiguous ///////////
1.5–0.1i	19	11	19	///////////
1.5–0.2i	26	20	37	
1.5–0.3i	27	24	49	
1.5–0.4i	26	26	55	——Warming——
1.5–0.5i	22	27	58	
1.5–0.7i	17	24	55	

* For an aerosol whose radii range from $r_1 = 0.01$ μ to $r_2 = 10\mu$.
† Ambiguous area is cross hatched.

"ambiguous" effect is decreased to $10^{-3} < n_2 < 10^{-2}$. While it is necessary to point out that climatological models of this sort clearly need to be improved and tested experimentally, it is also clear that they are *very* sensitive to the imaginary part of the refractive index. Aerosol absorption is probably the greatest for an industrial area with low humidity and may be least in remote humid locations, indicating a need for spatial studies as well.

The differences in the absorption-to-backscatter ratio for various size distribution exponents β depends on the relative fraction of particle sizes. Absorption and backscatter depend on the particle size for a given refractive index. A detailed description of the light scattering for absorbing particles has been reported by Plass (1966). In general, small particles ($r < 0.2 \mu$) backscatter a much greater fraction of the incident light (for Rayleigh scatters, one-half of the light is backscattered) than larger particles. The calculations for this reason may be conservative because the small size cutoff ($r = 0.01 \mu$) used in these calculations was the smallest claimed for the Junge distribution. Thus, the backscattering coefficient may be slightly larger than those calculated with different cutoffs or size distribution models. The light absorption coefficient for small opaque particles is approximately proportional to the volume of the particles (r^3), whereas larger particles absorb light proportional to the radius squared (r^2). The absorption of light also damps out oscillations in the scattering curves, the degree of which depends on the magnitude of the imaginary refractive index.

If the refractive index is measured for the atmospheric aerosol, the size distribution must also be determined in order to use this sort of calculation. Probably a more powerful approach would be to measure the integrated (b_{abs}/b_{bs}) ratio directly for an aerosol volume. This measured quantity could then be included in the more sophisticated climate models which can be expected in the future.

A considerable fraction of atmospheric aerosols may be composed of irregularly shaped particles. From studies of the scattering characteristics of irregularly shaped dusts, backscattering may be less than that predicted by Mie calculations for spherical particles (Holland and Gagne, 1970, Waggoner, 1971). This implies that our calculated ratios (b_{abs}/b_{bs}), which assume spherical particles, may be smaller those existing in the atmosphere.

5. Conclusions

Comparisons of the critical (b_{abs}/b_{bs}) ratio from earth-space heat transfer models and the (b_{abs}/b_{bs}) ratio for various aerosol models indicate that the magnitude of the imaginary part of the refractive index may cause changes in the temperature of the earth. Thus, cooling occurs for, $n_2 < 10^{-3}$ and heating for $n_2 > 10^{-1}$; the range $10^{-3} < n_2 < 10^{-1}$ is ambiguous.

It should be pointed out that, while the climate models are crude, they can probably be used to predict the critical absorption-backscattering ratio to within an order of magnitude, even though few if any measurements of this ratio have been made for atmospheric aerosols. Obviously, much more data on the light scattering and absorption properties of atmospheric aerosols on a worldwide basis are needed to determine if the secular increase of aerosol concentration from pollution will heat or cool the earth.

REFERENCES

Atwater, M. A., 1970: Planetary albedo changes due to aerosols. *Science*, **170**, 64–66.

Bryson, R. A., 1968: All other factors being constant. . . . *Weatherwise*, **21**, 56–61.

Bullrich, K., 1964: Scattered radiation in the atmosphere and the natural aerosol. *Advances in Geophysics*, Vol. 10, New York, Academic Press, 99–260.

Charlson, R. J., 1969: Atmospheric visibility related to aerosol mass concentration. *Environ. Sci. Tech.*, **3**, 913–918.

—— and M. J. Pilat, 1969: Climate: The influence of aerosols. *J. Appl. Meteor.*, **8**, 1001–1002.

Eiden, R., 1966: The elliptical polarization of light scattered by a volume of atmospheric air. *Appl. Opt.*, **5**, 569–575.

——, 1971: Determination of the complex index of refraction of spherical aerosol particles. *Appl. Opt.*, **10**, 749–754.

Fischer, K., 1970: Measurements of absorption of visible radiation by aerosol particles. *Beitr. Phys. Atmos.*, **43**, 244–254.

Hänel, G., 1968: The real part of the mean complex refractive index and the mean density of samples of atmospheric aerosol. *Tellus*, **20**, 371–379.

Holland, A. C., and G. Gagne, 1970: The scattering of polarized light by polydisperse systems of irregular particles. *Appl. Opt.*, **9**, 1113–1121.

Junge, C. E., 1963: *Air Chemistry and Radioactivity*. New York, Academic Press.

Landsberg, H. E., 1970: Man-made climatic changes. *Science*, **170**, 1265–1274.

McCormick, R. A., and J. H. Ludwig, 1967: Climate modification by atmospheric aerosols. *Science*, **156**, 1358–1359.

Mitchell, J. M., 1971: The effect of atmospheric aerosols on climate with special reference to temperature near the earth's surface, *J. Appl. Meteor.*, **10**, 703–714.

Plass, G. N., 1966: Mie scattering and absorption cross sections for absorbing particles. *Appl. Opt.*, **5**, 279–285.

Porch, W. M., R. J. Charlson and L. F. Radke, 1970: Atmospheric aerosol: does a background level exist? *Science*, **170** 315–317.

Report of the study of critical environmental problems, 1970: *Mans Impact on the Global Environment*. Cambridge, MIT Press, 88–91.

van de Hulst, H. C., 1957: *Light Scattering by Small Particles*. New York, Wiley, 114–130.

Waggoner, A. P., 1971: Measurement of the atmospheric aerosol at 694 mm. Ph.D. dissertation, Dept. of Geophysics, University of Washington, Seattle.

Wilf, H. S., 1967: Advances in numerical quadrature. *Mathematical Methods for Digital Computers*, Vol. 2, New York, Wiley, 133–144.