On the Impact of Pacific Ocean Free Tropospheric Background Aerosols at the Surface of the Earth-Atmosphere System

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RESUMEN

Se muestrearon partículas troposféricas individuales (0.03 a varias µm de diámetro) usando impactadores de alambre en un avión DC-864 a lo largo y dentro de la región del Pacífico de 125°E a 120°W y 70°S a 70°N, durante los desplegamientos de NASA del Experimento Global de Retrospección (GLOBE) en noviembre de 1989 y mayo de 1990. Fue usado un modelo de balance simple de radiación para determinar el efecto radiativo de primer orden de esta capa de aerosoles en la superficie.

Los resultados indican que: (i) los aerosoles troposféricos de fondo (espesor óptico de ≈ 0.05) entre 2.4 - 12.2 km ejercen una tendencia al calentamiento sobre superficies con albedo $> \approx 0.02$, tanto en el visible como en el infrarrojo. (ii) hay un aparente aumento en el modo tosco del sulfuro conteniendo abundancia de aerosoles comparado con \approx hace 15 años, sugiriendo que la capa de trasfondo de aerosoles ejerció una tendencia más fuerte de calentamiento planetario \approx hace 15 años

ABSTRACT

Individual tropospheric particles (0.03 to several μm in diameter) were sampled using wire impactors on a DC-8 64 aircraft along and within Pacific Rim, from 125°E to 120°W and 70°S to 70°N, during the November 1989 and May 1990 Global Backscattering Experiment (GLOBE) deployments of NASA. A simple radiative balance model was used to determine the first order radiative effect of this aerosol layer at the surface. The results indicate that: (i) The background (optical thickness of ≈ 0.005) tropospheric aerosols between 2.4 - 12.2 km exert a warming tendency on surfaces with albedos > ≈ 0.02 in both the visible and infrared. (ii) There is an apparent increase in the coarse mode sulfur containing aerosol abundance compared to ≈ 15 years ago, suggesting that the background aerosol layer exerted a stronger tendency of planetary warming ≈ 15 years ago.

1. Introduction

Clouds and atmospheric gases are well known to affect our climate (e.g. Dickinson et al., 1978; Wuebbles et al., 1989; Cohen, 1990; Wood, 1990; Oglesby and Saltzman, 1992). Twomey et al. (1984), Keopke (1992) and others report that the combined climatic effect of clouds and atmospheric aerosols is comparable to that of anthropogenic greenhouse gases but opposite in sign. However, very little attention (relative to clouds and gases) has been given to the climatic forcing by aerosols (WMO/WMP, 1992), perhaps due to the complexity of aerosol interactions and their influence on climate (Keopke, 1992). Aerosols are generally known to directly or indirectly affect the Earth's radiation balance leading to cooling or warming of the climate system. They directly affect radiative balance due to their reflection and/or absorption of radiation (visible and infrared). In contrast, they exhibit an indirect affect on the radiation balance through their influence on cloud formation, microstructure and duration, all of which affect cloud cover.

The warming or cooling of the surface by aerosol layers depends on the spatial and optical the properties of the aerosol layer, the altitude of the aerosol layer, the surface albedo and the zenith Research angle of the sun (Charlson and Pilat, 1969; Atwater, 1970; Mitchell, 1971; Grassl, 1973; Wang and Domoto, 1974; Coakley and Chy'lek, 1975; Reck, 1975, 1976; Charlock and Sellers, 1980; Reck and Hummel, 1981; Twomey et al., 1984; Coakley and Cess, 1985; MacCracken et al., 1986; Götz et al., 1991; Charlson et al., 1992; Peñaloza, 1996). The climatic effect of aerosol layers is estimated using models. The input aerosol layers most frequently included, layers of either (i) average or polluted aerosol concentrations, (ii) aerosols from volcanic eruptions (Snetsinger et al., 1987) to nuclear war, or (iii) carbonaceous Arctic aerosols (MacCracken et al., 1986). The background aerosol contribution, on the other hand, has received very little attention. Charlson et al. (1992) stress the importance of accounting for the climatic effects of aerosols in order to evaluate anthropogenic influences on past, present and future climate. Pueschel et al. (1994) report on a rare background tropospheric aerosol data set that provides an opportunity to gain some insight into the climatic forcing from background tropospheric aerosols, which is the objective of this study.

2. Background

1) Experimental details and some aerosol properties

Aerosol sizes and concentrations were measured with \approx 2 km resolution (average aircraft speed of 200 m s⁻¹) by an optical particle counter from Particle Measuring System, FSSP300 (Table 1; Pueschel *et al.*, 1994) and concurrently with ninety six (96) \approx 120 km resolution intervals by wire impactors (Farlow *et al.*, 1979) mounted on a DC-8 aircraft as part of the Global Back-Scattering Experiment (GLOBE) missions of NASA during November 1989 (GLOBE I) and May 1990 (GLOBE II). The sampling took place essentially along and within the Pacific Rim (125°E to 120°W and 70°S to 70°N; Fig. 1) in the free troposphere between 2.4 and 9.1 km mean sea level, and the upper troposphere between 9.1 and 12.2 km. All of the samples chosen in this analysis were obtained in cloud free air. The particle diameter obtained from the scanning electron microscopic (SEM) analysis of the wires ranged between 0.1 and 3.5 μ m, with a mean value of \approx 0.7 μ m for both campaigns. The chemical composition from electron microprobe X-Ray energy analyses (responsive to elements with atomic number, A > 10) and the diameter data are based on over 1225 individual particles distributed throughout the troposphere (2.4 - 12.2 km) over the Pacific Ocean Basin. The particles chosen for SEM/X-ray energy analysis were randomly selected based on the following assumptions; (a) uniformly distributed particles with respect to composition, size and

Table 1.	Optical properties of aero	sols in the troposphere over the Pacifi	ic Ocean Basin (Adapted from Pueschel et al., 1994).

Wavelength	Aerosol Extinction	Aerosol Backscatter	Aerosol Physical characteristics ^a		
(μm)	coefficient (km ⁻¹)	coefficient (km ⁻¹)	N_o (cm ⁻³)	D _g (μm)	$\sigma_{\!\scriptscriptstyle g}$
0.525 10.64	(2.03±1.20) x 10 ⁻⁴ (8.13±6.47) x 10 ⁻⁶	(6.45± 3.49) x 10 ⁻⁶ (9.98±10.69) x 10 ⁻⁸	20±20 20±20	0.12±0.02 0.12±0.02	3.0±0.2 3.0±0.2

The characteristics are the first component values of a two component lognormal fit of 22 samples collected through out the entire Pacific Ocean Basin. The minimum aerosol number concentrations of diameters, D>0.35 μm are typically between 0.16 and 0.25 cm⁻³, independent of time of sampling and of altitudes above 8.2 km.

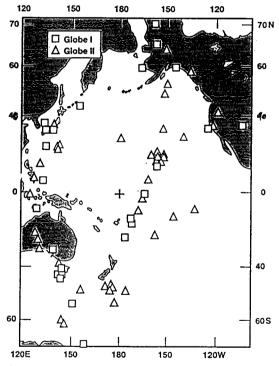


Fig. 1. Locations of the analyzed particulate samples from the two Global Backscattering Experiment (GLOBE) sampling campaigns, namely GLOBE I in November, 1989 and GLOBE II in May, 1990.

morphology in the 10 μ m interval bisected by the centerline of the exposed ≈ 2 cm long, 75 μ m diameter palladium wire and (b) the particles were representative of the numbers, types of particles along the horizontal altitude where they were obtained. According to Pueschel *et al.* (1994), (i) this tropospheric aerosol layer is horizontally homogeneous in its aerosol content (0.3 < D < 20 μ m) over hundreds of kilometers. Eventhough, the homogeneity of aerosols < 1 μ m diameter depends on their formation and removal mechanisms. (ii) the size distribution obtained from the SEM analysis corresponds very well with that from calibrated optical particle measuring probes. The chemical analysis is confined to the elements S.

Cl, Na, Si, and Mg. Other elements, however, such as Alet , P, Ca, K, Fe, Cu, Ni, were encountered in < 1% of total particles analyzed. A particle was considered to contain a specific element if the respective energy peak was at least 3 times the background (Bertin, 1963). The representative physical and chemical characteristics of the Pacific Ocean Basin Tropospheric aerosol particles, grouped according to diameters < 1 μ m (fine mode) and \geq 1 μ m (coarse mode), are shown in Figure 2 as a function of altitude. The composition within the sampled tropospheric aerosol layer appears to be approximately constant in the vertical.

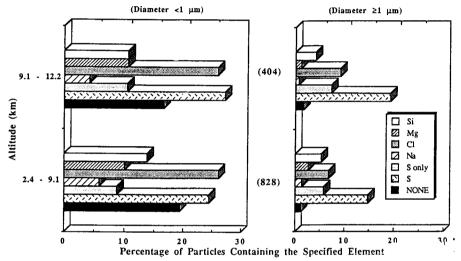


Fig. 2. The number fractions (in percent of total) for particles that contain elemental S, Cl, Si, Na, and Mg during GLOBE I and GLOBE II missions with respect to diameter. Particles composed of elements with A < 10 are denoted as NONE. The NONE group of particles may be indicative of nitrates and/or organic matter. They could also be particles with signals from elements of A > 10 that were not sufficiently above background level. "S only" indicates S presumably mixed with elements of A < 10, and is most likely of sulfuric acid, and/or other sulfates (e.g. ammonium sulfate). The value in parentheses is the total number of particles analyzed in the particular atmospheric layer. The percentage in any one layer exceeds 100% if more than one element is contained within at least one particle.

2) Simple radiative balance model

Since we are interested in the first order effect of an aerosol layer on the surface of the Earth-atmosphere system, consider a simple two layer system with (i) a homogeneous aerosol layer of reflectance, Re, and transmittance, Tr, (ii) an aerosol-free layer below it, and (iii) a surface with albedo, α , at the bottom of the aerosol-free layer. Furthermore, if we assume a thin aerosol layer, only one scattering event, and no clouds, then the balance between the radiative flux for this system without aerosols and the radiative flux of the aerosol layer may be expressed as;

$$\alpha = \text{Re} + [(\alpha * (\text{Tr})^2) * (1 - (\alpha * \text{Re}))^{-1}], \tag{1}$$

where, Re, Tr and τ (i.e., optical thickness of the layer) may be given by:

Re =
$$(\overline{\omega} * \tau)$$
- $(\overline{\omega} * g * \tau)$, (2)
Tr = 1 - τ + $(\overline{\omega} * g * \tau)$,

$$\tau = \int_{Z=Z_{bot}}^{Z=Z_{top}} E(\lambda, m)^* \sec \theta * dZ,$$

$$E(\lambda, m) = \int_{D=D_{\min}}^{D=D_{\max}} 0.25 * \pi * D^2 * Q_{ext}(D, \lambda, m) * \partial N * [\partial D]^{-1} * dD.$$
(3)

Where Z is the vertical distance (defined as positive in the upward direction); $E(\lambda, m)$ is the total extinction; $Q_{\text{ext}}(D, \lambda, m)$ is the MIE extinction efficiency factor at a given wavelength for a particle of diameter, D, and a refractive index, m; $\overline{\omega}$ is the single scattering albedo of the aerosol particles and is obtained from the ratio of the backscattering coefficient to the extinction coefficient; g is the scattering symmetry factor, with g = 1 for complete forward scattering and 0 for symmetric or isotropic scattering. Table 2 shows g and $\overline{\omega}$ for some aerosol types at wavelengths of 0.55 and 10 μ m based on the Standard Radiation Atmosphere in WMO/ICSU (1983). Pueschel et al. (1994) computed the light extinction and backscatter properties in the wavelength range 0.385 < λ < 10. 64 μ m using MIE theory for the background Pacific Basin aerosols.

Table 2. π and g for some aerosol types (after Standard Radiation Atmosphere in WMO/ICSU, 1983).

$\boldsymbol{arpi_{10}}^{\mathtt{a}}$	$\boldsymbol{\varpi}_{0.55}^{\mathbf{a}}$	g	$\beta_{10} / \beta_{0.55}^{b}$	
0.010	0.010	0.73	0.05	
0.000	0.209	0.34	0.038	
0.173	0.647	0.59	0.033	
0.130	0.947	0.70	0.035	
0.209	0.957	0.63	0.019	
	0.010 0.000 0.173 0.130	${f \varpi_{10}}^{a}$ ${f \varpi_{0.55}}^{a}$ 0.010 0.010 0.000 0.209 0.173 0.647 0.130 0.947	$egin{array}{cccccccccccccccccccccccccccccccccccc$	

a ω_{10} and $\omega_{0.55}$ are the single scattering albedo at $\lambda = 10.0$ and 0.55 μm respectively. b The ratio of the extinction coefficient at $\lambda = 10$ μm to the extinction coefficient at $\lambda = 0.55$ μm .

They assumed spherically shaped aerosols, as well as a refractive index between ammonium sulfate $(NH_4)_2SO_4$, and sulfuric acid, H_2SO_4 , to account for the prevalence of S in the particles. The particulate volume backscatter coefficients were calculated in a similar fashion. The corresponding characteristics at $\lambda = 0.525$ and $10.64\mu m$ are provided in Table 1. The optical depth, τ , of the tropospheric aerosol layer between 2.4 - 12.2 km over the Pacific Ocean Basin is ≈ 0.005 (Pueschel *et al.*, 1994), or background (Matsumoto *et al.*, 1987). Peñaloza (1996) calculated the ratio of the solar extinction to the infrared extinction for an atmosphere with only air molecules and obtained a value of 26. The equivalent value for

the GLOBE dataset (Table 1) is 25. The ratio of the solar extinction to the infrared extinction decreases as the visible optical depth of the layer increases, and so it is reasonable to consider the GLOBE data as representing background aerosols. Additional details of the aforementioned radiative transfer expressions may be found in Götz et al. (1991), Peñaloza (1996), and others.

3. Results and discussion

The effect of an aerosol layer on the net cooling or warming of the surface below may be assessed by comparing the reflectance and transmittance properties of the aerosol layer to equilibrium values for a particular surface with albedo, α . The solid lines in Figure 3a,b represent the surface albedo for which the insertion of an aerosol layer with reflectance, Re, and transmittance, Tr (or an increase in the particle concentration in an existing aerosol layer), has no effect on the albedo of that surface. That is, the solid lines represent equilibrium surface albedos. If one plots the Re and Tr of an aerosol layer on a graph in Figure 3 and the point lies to the left of any solid line, then this aerosol layer causes an initial tendency of planetary warming. Similarly, if the Re, Tr pair falls to the right of a solid line, then there is an initial

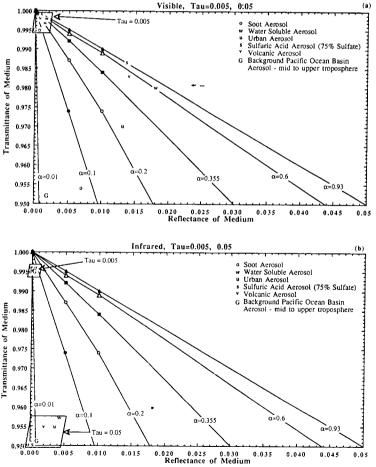


Fig. 3. The effect of an aerosol layer on the net cooling or warming of the surface given the aerosol layer transmittance and reflectance values (after Götz et al., 1991) for layers with optical thickness, Tau, = 0.005 and 0.05 at (a) a wavelength, $\lambda = 0.525 \,\mu\text{m}$, and (b) $\lambda = 10.64 \,\mu\text{m}$.

tendency of planetary cooling. For example, suppose a thin aerosol layer (optical thickness of 0.005) with Re = 0.001, Tr = 0.995 moves over a surface with albedo of 0.6. The Re, Tr point for this aerosol layer would be to the left of the equilibrium line for the surface with albedo of 0.6 (Fig. 3), and would consequently provide an initial tendency of planetary warming. But if the same aerosol layer moved over a surface with an albedo of 0.01, then the Re, Tr point would be to the right of the equilibrium albedo line for the surface with albedo of 0.01, and would consequently provide an initial tendency of planetary cooling. The letters shown in Figure 3 represent the Re, Tr pairs that correspond to the equilibrium surface albedo for the particular aerosol layer. Thus the line representing the equilibrium surface albedo for the particular aerosol layer could be drawn from Re = 0, Tr = 1 through the plotted letter down to Re = α , Tr = 0. Figure 3a indicates the effect of the inserted aerosol layer in the visible and Figure 3b in the infrared (IR) wavelengths. Typical reflectance and transmittance values of some aerosol types are shown in Table 2 and a few are used for comparison.

First the applicability of the simplified model (i.e., Eq. 1-3) is checked. The optical depths of the aerosol layers used here are 0.005 and 0.05, or 10 times greater than background. The volcanic aerosol layer with optical thickness of 0.05 gives an initial cooling tendency over surfaces with albedo, $\alpha < \approx 0.57$ in the visible (Fig. 3a) and over surfaces with $\alpha < \approx 0.02$ in the IR (Fig. 3b), which are qualitatively consistent with Götz et al. (1991). Sulfuric acid aerosol layers of optical depths 0.005 and 0.05 would result in initial cooling tendencies over nearly all surfaces in the visible including possibly those covered by clouds. This is consistent with Harshvardhan and Cess (1976). The sulfuric acid aerosol layers of optical depths 0.005 and 0.05 would lead to an initial warming tendency over all surfaces in the infrared. Harshvardhan and Cess (1976) also show a warming effect for a sulfuric acid aerosol layer in the infrared, but the combination of the visible and IR effects would still result in a cooling of 0.25 and 1.5 °K if the layer had an optical thickness, τ , of 0.005 and 0.05, respectively. Increasing the aerosol abundance (i.e. from 0.005 to 0.05; Fig. 3) of each aerosol type leads to cooling over additional surfaces as suggested by Twomey et al. (1984).

The background Pacific Ocean Basin tropospheric aerosol layer (BPOBTAL) is in approximate equilibrium with surfaces containing albedos of ≈ 0.01 , and would thereby exert an initial warming tendency over most surfaces in the visible and IR wavelengths (Fig. 3). When the optical depth of (BPOBTAL) is increased to 0.05, then (a) surfaces with albedo's greater than ≈ 0.02 would experience a warming tendency in the visible, and (b) virtually all surfaces would experience a warming tendency in the IR. Coakley *et al.* (1983), Coakley and Cess (1984) and others (Götz *et al.*, 1991) showed background aerosol to cool land and marine surfaces, especially at latitudes north or south of 70°. However, the optical thicknesses of the aerosol layers used by Coakley *et al.* (1983) and Coakley and Cess (1984) are > 0.07 and $> \approx 0.1$, respectively. These optical thicknesses are greater than that of the BPOBTAL, and based on Twomey *et al.* (1984) would be expected to yield a stronger planetary cooling tendency. The climatic forcing due to background aerosols at the surface is also related to the zenith angle of the sun, aerosol layer altitude, and chemical composition.

It is worth noting that the presence of sulfur (S) in both fine and coarse mode particles (Fig. 2) appears contrary to previous studies (Dzubay et al., 1977; Miller, 1979) that found sulfates, ammonium salts, soot and organic particles from condensed vapors in fine mode particles; Ca, Mg, K, Si, and Al in coarse mode particles, and chloride and nitrate in both modes. The NONE category of particles (those with no detectable elements, or with elements of atomic number smaller than 10) also exists in both modes and was

not explicitly mentioned in the previous studies. The discrepancy in S-containing species, especially, could be due to more accurate present day analytical techniques and volcanic eruptions (eq. Snetsinger et al., 1987). However, it might be due to a significant change in the composition of the larger aerosol fraction (Hoppel, 1988; Hegg, 1990, 1991; Hoppel et al., 1990). Given the latter, then it is reasonable to surmise that the background aerosol has increased in its sulfur content. A background aerosol layer with a higher sulfur content would be in equilibrium with a surface of higher albedo (Fig. 3), and would consequently exert a stronger initial tendency of planetary cooling. The latter suggests that the background aerosol exerted a stronger tendency of planetary warming ≈ 15 years ago.

4. Conclusions

A background free tropospheric aerosol data set recently obtained over the Pacific Ocean Basin has provided an opportunity to gain some first order insights into the climatic forcing from background tropospheric aerosols. The results indicate that:

- (i) Background (optical thickness of ≈ 0.005) tropospheric aerosols between 2.4 12.2 km are in approximate radiative equilibrium with surfaces containing an albedo of ≈ 0.02 , and otherwise exert an initial warming tendency on surfaces with higher albedos in both the visible and infrared.
- (ii) There is an apparent increase in the coarse mode sulfur containing aerosol abundance compared to \approx 15 years ago, suggesting that the background tropospheric aerosol layer exerted a stronger tendency of planetary warming \approx 15 years ago.

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