Aerosols at Mauna Loa: Optical Properties

GLENN E. SHAW

Geophysical Institute, University of Alaska, Fairbanks 99701

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ABSTRACT

The spectral attenuation of sunlight passing through the atmosphere was determined with the Langley method for 110 clear days and at 11 wavelengths to an accuracy of $\delta\tau = \pm 0.002$ (τ is the optical thickness) at the Mauna Loa Observatory, Hawaii. Suspended aerosols above the observatory attenuated light by an average of 1.9% (in the vertical direction) at a wavelength of 5000 Å, and the average attenuation varied with wavelength as $\lambda^{-1.6}$. Air masses from northerly directions were most turbid, $\bar{\tau} = 0.021 \pm 0.015$, while those from southwesterly direction were least turbid, $\bar{\tau} = 0.017 \pm 0.005$. The lowest values of optical extinction varied as $\lambda^{-1.3}$ while those from directions of nearest continents varied as $\lambda^{-2.5}$; the larger values of wavelength exponent for the continental aerosol is what would be expected for an aerosol cloud that had been carried by the winds from distant continents. It is deduced that aerosol from North America and Asia occasionally reaches the Hawaiian Islands. The explosive eruptions of Augustine Volcano in Alaska (January 1976) caused a perturbation of $\delta\tau \approx 0.01$ at $\lambda = 500$ Å on the optical extinction and decayed with a time constant of five months.

1. Introduction

Mauna Loa Observatory, Hawaii, in the central Pacific, more than 3500 km from nearest continental land and at an elevation of 3380 m (11 100 ft), is an important benchmark climatic observatory where meteorological elements and trace constituents in the atmosphere are being continuously monitored. The purpose of this paper is to report on the characteristics of the vertically integrated aerosol over Mauna Loa. The aerosol was studied by carrying through a series of precision multi-wavelength photometric measurements of the sun's intensity to deduce the spectral extinction of the atmospheric aerosol and its wavelength-dependence. Seven months of data covering 110 clear days are being reported.

2. Mauna Loa Observatory—Topography and aerosol climatology

The observatory is on the north side of the gently sloping volcano, across from Mauna Kea Volcano; a saddle at an elevation of 2100 m separates the two mountains. There is no vegetation nearby and the surface is composed of volcanic rock containing practically no fine dust. Mauna Loa Observatory is upwind from the other islands in the Hawaiian chain.

A marine trade inversion traps particulates emitted by the ocean and the island, except near noon when the inversion breaks and air occasionally flows up the saddle to the observatory. When this happens local aerosol concentration (measured with nephelometers and Aitken particle detectors) increases (Charlson *et al.*, 1974) and fluctuates, but during subsidence in the evenings and morning hours, nephelometer and Aitken count readings remain small and constant. Nephelometer scattering during clean conditions at MLO is only about 7% above pure Rayleigh scattering (at 5000 Å).

Mendonca and Pueschel (1973) report concentrations of 0.5 ice nuclei per liter and 0.6 $\mu g\ m^{-3}$ of total atmospheric aerosol during downslope flow conditions. Ice nuclei counts go up by about a factor of 4 and aerosol mass loading goes up by a factor of 10 at times of well-developed upslope flow. Occasionally, slight disturbances in the particle readings occurs during downslope flow, but these are relatively infrequent. The background aerosol, as sensed at MLO, is probably representative of average conditions over the central Pacific Ocean for altitudes above the trade wind inversions.

3. Experimental technique

As early as 1721 (Middleton, 1961) the French physicist Jean Jacques de Mairan remarked that one could derive the opacity of the atmosphere, at least in principle, by measuring the relative intensity of sunlight or starlight at two different elevation angles. Use of the technique to derive turbidity is well known by now and is called the Langley method, after Samuel P. Langley who first fully developed it at the turn of the century. The Langley method is an empirical fit to the Lambert-Beer law which, when written logarithmically, has the form of a linear equation

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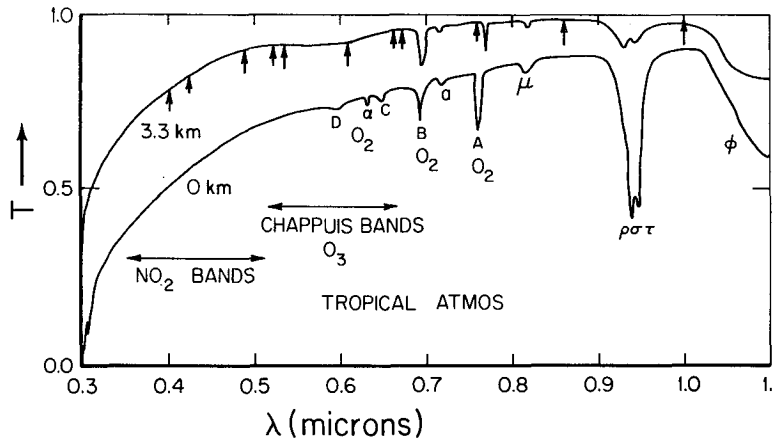

FIG. 1. Vertical transmission through the atmosphere for conditions at sea level, and at the altitude of Mauna Loa (3.3 km). Bands due to water vapor and oxygen absorption are indicated by their spectroscopic designation and the arrows point to wavelengths used in the experimental determinations of spectral extinction.

relating $\ln T$ and m_j:

$$-\ln T(\theta,\lambda) = \underset{\text{(molecular scattering)}}{m_1\tau_1} + \underset{\text{(ozone)}}{m_2\tau_2} + \underset{\text{(aerosol)}}{m_3\tau_3}, \quad \tau=\tau(\lambda), \quad m=m(\theta) \qquad (1)$$

where T is the slant-path monochromatic optical transmission through the atmosphere; τ_i is the optical thickness or the integral of the optical extinction coefficient for a ray traversing *vertically* through the atmosphere; m_i, the relative air mass, is a geometrical term necessary to account for the relative increase in optical path length when solar rays enter the atmosphere at elevation angle θ. The relative air mass very nearly equals the cosecant of the sun's elevation angle except at low elevation angles ($<15°$) when the curvature of the atmosphere makes it necessary to bring in correction terms and take into account the vertical distribution of the absorbing-scattering gases or particles. For such low-angle conditions the m_i are in general somewhat different, but the departure from $m_1=m_2=m_3=\text{cosecant}\theta$ is small enough to ignore provided $\theta>20°$.

Eq. (1) is used to determine optical depth τ at different wavelengths (which were selected by interference filters) by regression fitting many measurements made throughout a day of the sun's monochromatic brightness. Each day's data then provide an independent calibration of the instrument (in terms of the sun's extraterrestrial brightness), which was kept track of for the 110 days of Langley plot analyses made during this study. In this way it was found that the photometer drifted by less than 1% over one year's time. This gave the basis to be able to go back to individual data points and recover a very accurate (to ± 0.002) estimate of the optical depth at any specific time. This made it possible to determine short-term variations in the temporal behavior of optical depth.

In the visible optical spectrum, between roughly 4000 and 10 000 Å wavelength, the Langley-Beer law holds up at all wavelengths except in gas absorption features (see Fig. 1). Radiation in even weak absorption bands may depend on temperature, pressure and nonlinearly on the amount of absorbing material. Wavelengths used in this study were purposefully placed *between* H_2O and O_2 gas absorption features. One exception was a wavelength band at 1 μm, that consistently showed anomalously large optical extinction. This has been attributed to a weak water vapor continuum between the $\rho\sigma\tau$ and ϕ band systems.

The two major processes acting to diminish the intensity of incoming parallel monochromatic radiation are Rayleigh scattering and scattering and absorption by the suspended particles in the air. Ozone and nitrogen dioxide (Shaw, 1976) have weak continua in the visible which obey Beer's law (Inn and Tanaka, 1953; Hall and Blacet, 1952).

In this study the measurements of spectral extinction were made within 11 100 Å wide bands placed between gas absorption features shown in Fig. 1 and spanning the wavelength region $4000<\lambda<10\,000$ Å. In these specially selected regions Eq. (1) is valid. The residual component of absorption-plus-scattering by the atmospheric aerosols (τ_3) forms the main topic of interest in this paper.

With regard to the accuracy achievable with the Langley method in determining τ, it depends on the validity of the following assumptions: (i) the Lambert-Beer law is applicable, (ii) the atmosphere is horizontally homogeneous, (iii) the extraterrestrial solar irradiance is constant during measurements, (iv) the aerosol concentration is constant, (v) the measurement are monochromatic or nearly monochromatic, (vi) the instrumental sensitivity does not change, and (vii) diffuse radiation in the instrument's field of view is negligible in comparison to the direct solar beam.

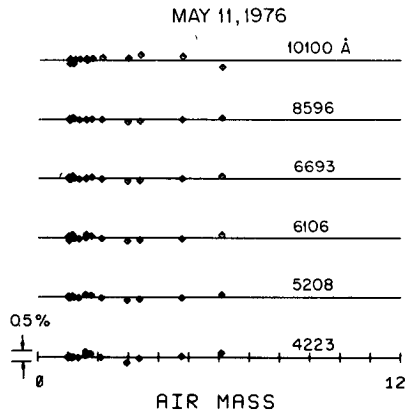

FIG. 2. Vertical departures of points about a least-squares-derived Langley line. An air mass of 1 corresponds to the sun at the zenith; an air mass of 12, to the sun at 4.7° elevation angle. The observed solar intensities (symbols) agree with those calculated with Eq. (1) to a few tenths of 1%. The size of a deviation of 0.5% is shown at the left of the bottom line.

These assumptions held to excellent approximation at Mauna Loa. The numerical values of τ being reported are estimated to be accurate to ± 0.002.

Assumptions (iv), (vii) and possibly (ii) usually are met only poorly at most continental locations and at some high-altitude observatories (Ångstrom, 1970; Russell and Shaw, 1975). This has caused innumerable problems with some of the past reported values of atmospheric spectral transmission!

The photometer used for the measurements at Mauna Loa was a filter wheel instrument having 12 well-blocked interference filters (out of band rejection $>10^{-6}$) and a solid state PIN-doped silicon photodetector, chosen for its proven stability. The instrumental field of view was 3° diameter. In operation, the output signal of the photometer was recorded to an accuracy of 0.1% with a digital voltmeter. Tests defined the temperature coefficients, the filter leakage and the overall stability of the instrument (which was ~1% per year). Measurements were taken at Mauna Loa every clear day from March to August 1976 and in January–February 1977.

4. Magnitude of the spectral extinction coefficients

Deviations about the least-square-fitted Langley plots were very small at Mauna Loa, strongly implying, but not proving absolutely, that the assumptions made in the Langley method held up well in practice. Typical deviations found in the quantity $\ln T - \ln T$ (Langley) are illustrated in Fig. 2; the rms deviation about the straight line for any given day's data was less than 0.5% for 80% of the data, and 30% of the time was only 0.1%. Thus the indications are, from the Langley plots, that one can assign high reliability to the slopes of the lines and hence to the derived values of spectral optical extinction.

The optical thickness of aerosols-plus-ozone was estimated at each wavelength band by subtracting the Rayleigh-scattering optical thicknesses from the derived optical thickness of the atmosphere above Mauna Loa. There is a certain amount of uncertainty associated with this process because the molecular scattering depolarization factor is not known perfectly; values of Rayleigh-scattering optical thickness calculated for a tropical atmosphere and reported by Fröhlich and Shaw (1978) were used for the calculations. These agreed to three decimals with values calculated by Hoyt (1977). They were 3.6% lower than those listed by Valley (1965).

It was impossible to avoid the weakly absorbing Chappuis ozone bands, since they are so broad, and the aerosol extinction spectrum therefore contains a residual component due to ozone absorption. This was subtracted out in the final stages of the analysis to give an estimate for the aerosol extinction spectrum, i.e., the wavelength dependence of aerosol extinction. The aerosol extinction was small at Mauna Loa, about an order of magnitude less than at most rural continental locations (Flowers and Viebrock, 1968).

Fig. 3 shows some examples of aerosol-plus-ozone extinction spectra, while the frequency of occurrence distribution of the aerosol-plus-ozone optical thickness for 110 clear days at Mauna Loa is shown in Fig. 4. After calculating and removing ozone absorption and absorption from an estimated 1×10^{-3} cm STP NO_2 (Shaw, 1976), the mean aerosol spectral extinction at Mauna Loa was 0.019 ± 0.010 at 5000 Å wavelength

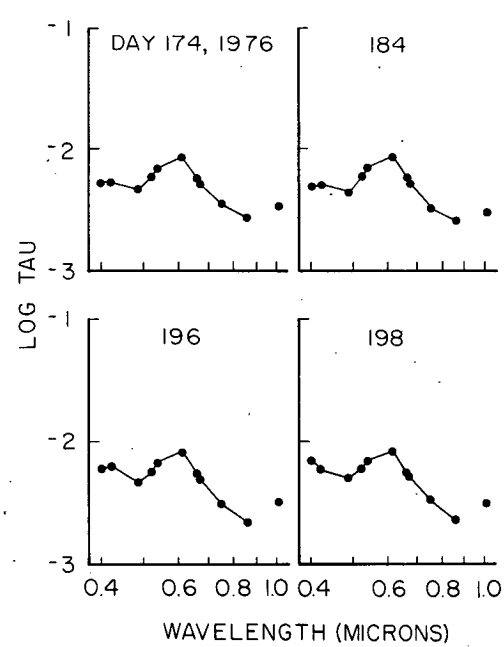

FIG. 3. Example plots of the wavelength-dependence of log optical thickness (log τ) for combined aerosol-plus-ozone extinction. The anomalously high extinction at 1.01 μm is caused by absorption in a weak water vapor continuum between the $\rho\sigma\tau$ and ϕ water vapor bands. The ozone Chappuis bands show as a bump in the extinction spectra peaking at 6000 Å.

and varied with wavelength approximately as $\lambda^{-\alpha}$ with $1.1<\alpha<3.5$, but averaging $\alpha=1.63$.

5. Stratospheric aerosol from Augustine Volcano eruption

The spread of the frequency of occurrence distribution in Fig. 4 is to a large extent caused by a slow decrease in τ_3 (Fig. 5) that occurred from March to August 1976 and which was possibly due to the decay of stratospheric aerosol resulting from a series of eruptions of Augustine Volcano (59.36°N, 153.43°W) in the Gulf of Alaska in January and February 1976. Unfortunately, observations at Mauna Loa began in early March just after the Augustine eruptions, so it could not be verified that the decay was caused from the volcano, but no other major eruptions occurred in late 1975 or early 1976. If the Augustine Volcano eruptions caused the excess extinction at Mauna Loa, then the maximum excess optical thickness in the stratosphere in early March (two months after the eruptions) was $\delta\tau=0.010\pm0.005$ at $\lambda=5000$ Å and followed approximately a $\lambda^{-1.3}$ relationship. The decay of material from Augustine was exponential with time with a time constant of 5 ± 2 months. If it is assumed that the stratospheric veil from Augustine covered one-third of the earth's surface, the Dust Veil Index (DVI; Lamb, 1970) would only be in 2 comparison with DVI=1000 for the Krakatoa eruption in 1883; DVI=800 for the Agung eruption in 1963 and DVI =150 for the Katmai eruptions in 1912. It is remarkable that relatively small eruptions like the Augustine eruptions could actually generate enough material in the stratosphere to be detected at Mauna Loa. Apparently it would be practical to keep a watch on volcanic activity by making continual measurements of atmospheric spectral transmission from high-altitude

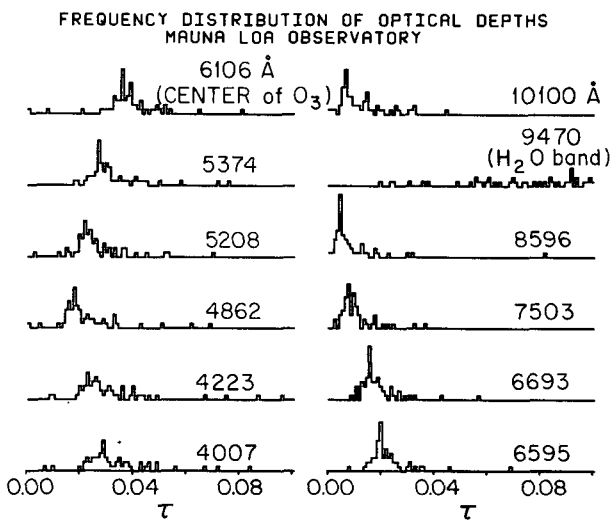

FIG. 4. Frequency of occurrence distribution for aerosol-plus-ozone optical thicknesses from slope of Langley plots for 110 clear days at Mauna Loa.

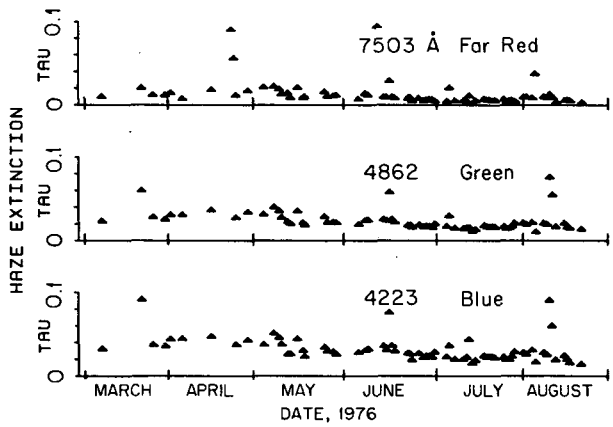

FIG. 5. Temporal variation in aerosol spectral extinction at three wavelengths from 15 March to August 1976. The decay of the stratospheric veil from Augustine Volcano, which erupted in January and February 1976, is attributed to the decaying optical thickness. The six high points were contaminated with thin barely-visible cirrus.

observatories. All further information reported in this paper is on the observed values of τ *minus* the exponentially decaying τ's from the volcano. This should pertain to the natural aerosol above the trade wind inversion in the central Pacific ocean.

6. Diurnal variations in optical thickness and relations to station meteorology

Several typical patterns of diurnal variations of optical thickness above the observatory are shown in Fig. 6. Occasionally the turbidity remained constant during the day, but in 70% of the cases studied it increased in the afternoon hours. In general, the turbidity was smallest and most constant during the early morning hours when solar elevation was less than about 60°. The upslope winds which form in the afternoon hours apparently contain enough contamination to cause the optical thickness to increase by as much as $\tau_3=0.01$, which, assuming an aerosol of 50 μg m^{-3} mass load, would correspond to a laminar upflowing layer thickness of ~100 m. As mentioned, the effect of upslope contamination was also apparent in condensation nucleus count and in the nephelometer measurements at Mauna Loa. The conclusion is that the most meaningful background turbidity values come from the morning sun photometer measurements.

There was no apparent relationship between humidity at the station and the values of τ_{5000}. The diagram in Fig. 7 shows the day-averaged spectral optical thickness at 5000 Å wavelength plotted against the station's nephelometer readings. There is a slight hint in Fig. 7 that τ_{5000} may correlate with the nephelometer readings. This would suggest that part of the aerosol sampled by "point" sampling experiments at the station is responsible for the optical extinction. No relation between these variables would be expected

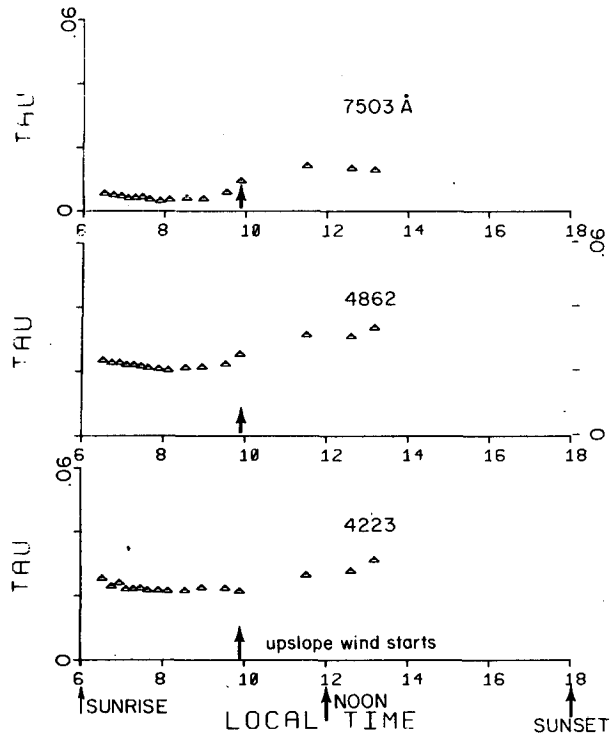

FIG. 6. Example of diurnal variation of the aerosol optical thickness τ above the observatory. The arrow marks the time when contaminants, carried up the slopes of Mauna Loa by anabatic winds, were detected at the observatory.

if the majority of aerosol optical thickness was contained in a semi-permanent stratospheric cloud. No relation was found between turbidity and the station's temperature and wind speed, at least at times when the air was not contaminated by upflowing winds.

7. Relations between spectral extinction and back air trajectories

Air trajectory information supplied by Dr. John Miller, Director of the Mauna Loa Observatory, was related to spectral turbidities for each day's data taken from March to August 1976. The air trajectories were computed, going back 10 days from $t=0$, and grouped in classes according to the distance (500, 1000, 1500 km) and azimuth sectors (defined by five segments in the SE, NE, N, NW and SW directions) reached. A comparison of air trajectories with the optical extinction parameters (Table 1) shows essentially no correlation between *distance* and turbidity, but τ and the Ångstrom coefficient α (where $\tau = \text{constant} \cdot \lambda^{-\alpha}$) show a dependence on *azimuth;* with the highest values of spectral extinction being associated with air masses coming from the N and NE and the lowest values with air masses originating from the SE and SW. The implication is that part of the aerosol extinction above Mauna Loa is caused from aerosols transported in from the direction of North America. The nearest points of continental land from Hawaii are the California coasts to the northeast (3600 km) and the Alaskan and northwest British Columbia coasts to the N and NE, both at ~4200–4400 km distance. Typical rural continental turbidities are ~0.1–0.2. The excess turbidities observed at Mauna Loa when the wind had been from continents was around 0.007, a factor of 14–28 times lower than turbidity over *rural* continental areas.

The southwest sector direction from Mauna Loa has the lowest turbidity values, a fact not suprising when one considers that Australia, the nearest continent southwest of Hawaii, is 8000 km distant, or twice as far away as the west coast of the United States and across the Intertropical Convergence Zone, which would scavenge particles out that attempted to pass through it.

Comparing the range of variation of the turbidities from different directions indicates that the maximum

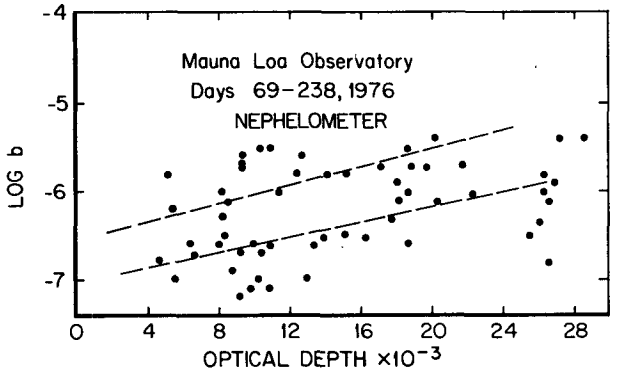

FIG. 7. Log b, the station's nephelometer scattering coefficient, plotted as a function of the aerosol optical thickness above the observatory.

TABLE 1. Average values and standard deviations of aerosol optical thickness at 5000 Å wavelength τ_{5000} and the Ångstrom wavelength coefficient α for mid-tropospheric 10-day back trajectories. Optical thicknesses have been multiplied by 1000.

	Direction*				
	SE	NE	N	NW	SW
τ_{5000}	18.3	20.8	24.3	19.5	16.9
$\sigma\pm$	12.8	14.7	12.3	9.7	5.0
α	1.51	2.03	1.72	1.27	1.28
$\sigma_\alpha\pm$	0.36	0.62	0.49	0.34	0.34

	Distance (km)		
	500	1000	1500
τ_{5000}	18.7	19.2	21.8
$\sigma\pm$	12.0	11.9	11.6
α	1.50	1.75	1.58
$\sigma_\alpha\pm$	0.44	0.66	0.43

* Air flow from.

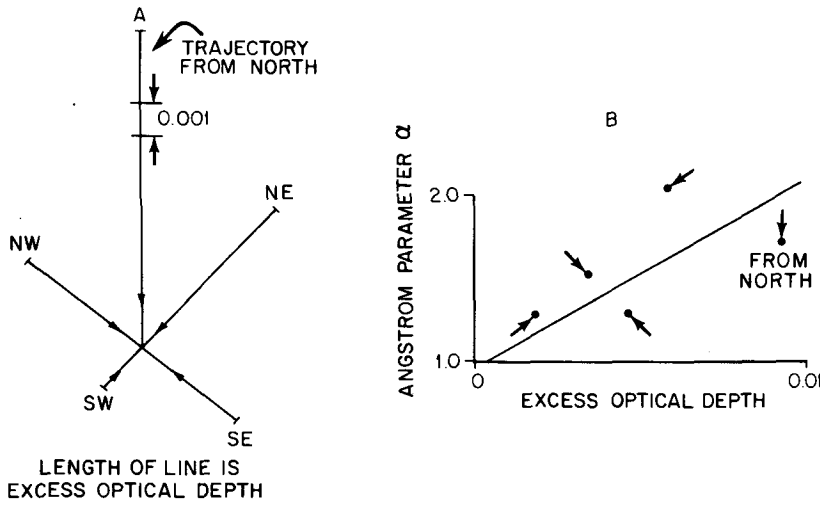

FIG. 8A. Variation of estimated excess aerosol optical thickness (observed minus 0.015) above the observatory for different air mass origins 10 days before the observations. Air originating from northern directions is most turbid, while that from southwesterly directions is least turbid. The length of the vectors are proportional to the average values of excess optical depth; the length of an increment of $\tau=0.001$ is marked.

FIG. 8B. The average value of the Ångstrom coefficient $\alpha(\tau=\text{constant}\cdot\lambda^{-\alpha})$ plotted against excess aerosol optical thickness. The five points refer to air mass origins from five different directions (shown as arrows). A trend of increasing α with increasing τ is illustrated by the solid line.

aerosol optical extinction from continental aerosols carried by the winds to Hawaii ($\tau=0.007$) is about one-third of the total average aerosol extinction (0.019); the remaining component is probably global and almost certainly consists partly of coagulated products of photochemically nucleated aerosol from trace gases such as sulfate particles or converted hydrocarbons. It appears that this general background aerosol over the Pacific, from whatever its source, causes an additional average vertical optical extinction for downward-directed rays of around 1.3% at 5000 Å wavelength.

8. Discussion

It is interesting to compare the "continental component" of the aerosol optical extinction with the Ångstrom wavelength exponent α. The continental component is defined by subtracting 0.015 (somewhat smaller than the average optical extinction from the cleanest azimuth) from the time-averaged values of observed aerosol extinction for different air trajectory directions. The results of the subtraction to give the continental component are shown in Fig. 8 where it is seen that higher values of continental aerosol extinction seem to be associated with higher values of wavelength exponent α. This implies that the background aerosol cloud from the cleanest directions has a value of α around unity, in agreement, incidently, with the optical extinction computed for the stratospheric aerosol size distributions listed by Bigg (1976). Fig. 8B shows that the continental-derived aerosol over the Pacific (for airflow from the north) gives rise to an aerosol optical extinction spectrum with larger α; in other words, the optical depth of the transported, but aged, continental aerosols falls off more rapidly with increasing wavelength than the background aerosol from the cleanest directions.

The rapid falloff of aerosol optical thickness with increasing wavelength is hypothesized to indicate the presence of an aged tropospheric aerosol. The main empirical evidence for this are the data of Misaki *et al.* (1975) who studied the deformation of the size distribution of aerosols as they evolved during transport out to sea to distances of 1000 km from Tokyo Harbor. Calculations made by the author (unpublished at present) on the wavelength dependence of the optical extinction through such an evolving cloud of particles (using data from Misaki *et al.*) showed a persistent tendency to give steeper falloffs of optical depth with wavelength with increasing age. The result was always the same and almost independent of the size distribution of the original starting particles. It seems almost certain that aged air masses have steeply falling aerosol size spectra or, equivalently, relatively large values of the Ångstrom parameter α.

The hypothesis that excess optical extinction at Mauna Loa is due to distant sources of tropospheric aerosol is supported by the observed values of α, which are high as would be expected for an aerosol cloud transported for long distances in the troposphere. The enhancement in spectral extinction from

the continental aerosol can provide a rough estimate of the mass loading and number concentration of particles in the air column above the observatory. The extinction spectra followed a $\lambda^{-\alpha}$ law. This sort of wavelength dependency indicates that the aerosol size spectrum dn/dr probably closely followed a $r^{-\beta}$ relationship, where $\beta \approx \alpha+3$ (Shaw *et al.*, 1973). As mentioned, the "turbid" air flowing from continents had a relatively large value of α, between 2 and 4. Adopting $\alpha=3$ and $\Delta\tau=0.007$, and assuming that all particles are in the size range $0.05\ \mu m < r < 1\ \mu m$, the mass loading of continental-derived material (a particle density of 1 g cm^{-3}) above the station is 5×10^{-7} g cm^{-2} or, if the particles are uniformly distributed throughout a 5 km thick column, the point mass loading would be 1×10^{-12} g cm^{-3} or 1 μg m^{-3}. A monodisperse aerosol of 100 particles cm^{-3} and radius 2.3×10^{-5} cm would provide this particle load.

9. Conclusions

The results of this study can be summarized as follows:

1) Spectral extinction at Mauna Loa decreased from March to June 1976, probably as the result of the decay of a stratospheric dust veil caused from eruptions of Augustine Volcano, Alaska, in January–February 1976. The Dust Veil Index for the Augustine cloud based on the optical extinction data from Mauna Loa, would be only $\sim$2. The optical thickness of the stratospheric veil was 0.01 ± 0.005 (at $\lambda=5000$ Å) in early March 1976. Dust from the subpolar latitudes of injection reached the tropics in less than two months time and decayed with a time constant of five months. The stratospheric cloud's spectral attenuation varied approximately as $\lambda^{-1.3}$.

2) The optical thickness of aerosols at Mauna Loa is small and constant during morning hours, but increases and fluctuates in the afternoon from aerosol material that is carried up through the trade mixing layer by convection and anabatic (upslope) winds. The upflowing contaminants increase Aitken nucleus counts and nephelometer optical scattering. Most afternoon data were suspected of contamination and therefore were rejected for analysis in regard to studies of the background aerosol at MLO.

3) Evidence was found for the occasional incurrence of "continental aerosol" at Mauna Loa when mid-level tropospheric winds came from the direction of North America. The increase in optical thickness was $\sim$0.007 (at 5000 Å).

4) The apparent natural background aerosol at Mauna Loa (the lowest values of turbidity) had an optical thickness of $\sim$0.015 (5000 Å). These lowest values occurred when the winds came from southwesterly directions.

5) The aerosol extinction spectrum could be approximately expressed by a power law function of the form $\tau=\beta\lambda^{-\alpha}$. The stratospheric volcanic dust veil from Augustine had a value of $\alpha\approx1$–1.5, while the enhanced aerosol optical thickness from northern, northwestern and northeastern directions gave rise to steeper optical extinction spectra, with α sometimes exceeding 2. The higher values of α for the more turbid air supports the hypothesis that the excess aerosol consists of particles that had been carried in the middle troposphere from sources in Asia and North America.

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