

Background aerosol light scattering characteristics: nephelometric observations at Mauna Loa Observatory compared with results at other remote locations

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ABSTRACT

Background aerosol may be defined as that which exists in remote locations, well away from local variable sources. The total contribution to the optical aerosol extinction coefficient due to scattering and that contribution due to the scattering into the backward hemisphere (hemispheric backscatter) are important due to their role in determining atmospheric extinction and albedo respectively. Specially adapted instruments are described for measuring these parameters in remote sites at various altitudes in the troposphere. Preliminary field measurements are described and data compared for several sites and altitudes. The Mauna Loa Observatory, Hawaii, is of particular interest since it has been used for studies of CO₂ increases. The measured light scattering coefficient due to aerosol at 500 nm wavelength is ~10 % of the Rayleigh scattering of the gas molecules and provides a base line to which future changes may be referred. The results for the hemispheric backscatter (or albedo) indicate a value occasionally above that coefficient predicted by the Mie formulae and assumed size distribution of spherical particles.

I. Introduction

A. Rationale

In recent years, interest in a secular increase of the *background aerosol* has developed, largely due to speculation regarding its effects on regional or even global scale climate. The main concern seems due to the possibility that aerosol might be responsible for the cooling trend noted globally in meteorological data in recent years (Mitchell, 1971, 1972). Simultaneously interest has arisen regarding background levels of a variety of trace substances, including aerosol, for purposes related to air pollution control strategies. It is obviously necessary to know the composition of air in its natural state against which pollution can be measured and to which controls may bring air quality.

In both of these areas of interests, there have arisen controversies due to a lack of data on

relevant properties of particulate matter in the atmosphere in remote locations. Even if the relatively simple properties of mass or number concentration were known in such locations, the climatologically important radiative effects would not be easily understood. Such radiative effects may be segregated into *direct* and *cloud related* cases. The latter is due to the influence of the aerosol on cloud formation or on the optical properties of the clouds into which the aerosol is incorporated. Indirect aerosol effects could ensue due to dynamical feedback between the radiation field, atmospheric motions and clouds (Schneider, 1972).

In the simplest case of direct radiative effect of aerosol without cloud involvement, disagreement exists on even the sign of temperature change to expect from secular changes on regional to global scales.

Charlson & Pilat (1969) pointed out that the backscattering and absorption characteristics of atmospheric aerosol are not determined well enough to predict a heating or cooling trend due

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to future changes in aerosol loading in the atmosphere. Schneider (1971, 1972) and Rasool & Schneider (1971) reported that aerosols would uniquely cool the earth's surface given one set of assumptions which were disputed by Charlson & Pilat (1971) and by Charlson et al. (1972). In order to clarify this and related controversies, it seems necessary to carefully study the appropriate properties of aerosol in representative baseline sites (Munn & Bolin, 1971).

The main purpose of this paper is to report observations of some of the relevant optical properties of non cloud aerosol in remote locations. Ultimately these and other data should be useful in constructing less ambiguous models of the radiative effects of atmospheric particulate matter. However, before proceeding, it seems useful to briefly discuss what is meant by *background aerosol*.

B. Definition of background aerosol

Perhaps the first definition of *background aerosol* was made by Junge (1963) based on measurement of the Aitken or condensation nucleus (CN) count. This definition is restricted to upper tropospheric situations where the CN count of ca. 200 cm^{-3} is not influenced markedly by any sources. Junge also defines a *marine aerosol*, a *continental aerosol*, with variable CN counts and a *stratospheric aerosol*.

Other measures of aerosol properties in remote locations also have been used to define properties of *background aerosol*. Mass concentration, size distribution, chemical composition and various optical properties have been used.

Ultimately data on a wide variety of aerosol properties will clarify and refine the definition of *background aerosol* in various types of locations. Meanwhile, we shall define *background aerosol* simply as that which exists in remote locations, far from local sources, but may include the contribution from distant human activities. While this differs slightly from the original definition by Junge (1963), we feel the term background is most commonly used to imply a non-urban or remote situation rather than the single case of the upper troposphere.

Porch et al. (1970) suggested that a property of background particulate matter could be inferred from measurements made with a version of the integrating nephelometer originally devised by Beuttel & Brewer (1949). Specifically, Porch et al. (1970) measured the scattering com-

ponent of the extinction coefficient at sites where local aerosol sources were absent, such as at observatories on snow covered mountains or in the Arctic in winter. They hypothesized that the background aerosol is further characterizable by the small degree of variation in its scattering coefficient as a function of time.

A second purpose of this paper is to test this hypothesis by comparing data from the more or less accepted baseline site at Mauna Loa Observatory with other likely sites. It must be recognized that few quantitative or even qualitative criteria have been applied and tested in a systematic way toward the establishment of sites at which baseline values of relevant optical or other aerosol parameters may be measured as function of time. Munn & Bolin (1971) report a list of criteria suggested for such sites and it is hoped that these data will help to confirm their utility.

C. Secular trends and need for monitoring

There have been numerous attempts at estimated global aerosol budgets (Robinson & Robbins, 1971; Hidy & Brock, 1971; SMIC, 1971), all of which conclude that the human contribution is currently in the range between about 5 and 50% including all particle sizes and lifetimes. Since most human activity injects material into the lower troposphere over a small percentage of the area of the northern hemisphere and since the lifetimes of particles in the troposphere is short compared to the time to mix the troposphere, it seems likely that the current and future aerosol concentration maxima will be located in the vicinity of the sources. Scales of the order of 1 000 km seem to be most likely if an average residence time of 2–10 days is used (Rodhe & Grandell, 1972). Thus, unlike CO_2 which is well mixed in the whole atmosphere, the effects of human contribution to the aerosol budget should be much more localized. Further, the kinds of meteorological or climatological effects may be expected to be different than for globally distributed pollutants. Selective alteration of synoptic features in or near populated regions might occur due to patchy radiative or cloud related effects that would not occur if the aerosol were evenly distributed over the globe.

This spatial inhomogeneity may be expected to intensify due to the forecasted increase in anthropogenic aerosol. However, it is certain at this point that the spatial distribution of most

of the relevant aerosol properties are essentially unknown on scales of hundred to thousands of kilometers. To be sure, condensation nuclei count (Mohnen, 1973), mass concentration (Ludwig et al., 1970) and turbidity (Flowers et al., 1969) have been measured on such scales; however, none of these are known to be related in any simple way to weather and climate effects, and the latter two measurements may have such large biases that little confidence can be placed in them for detection of trends at remote sites (Whitby & Liu, 1970; Laulainen & Taylor, 1973) respectively.

The clearcut forecast of human domination of aerosols on regional scales has resulted in a consensus that appropriate monitoring of these substances is needed (SMIC, 1971). However, there is at the same time a lack of agreement on what quantities should be measured, what instruments should be used or where and how often measurements should be made. The large number of possible methods coupled with the largely unknown nature of the aerosol and its spatial distribution precludes an *a priori* approach toward establishment of monitoring systems. Rather, experimental measurements of a wide variety are needed in order to gain the insight needed for design of an adequate regional to global scale non-urban network for climatic monitoring.

The third purpose of this paper is thus hopefully to aid in establishing a sound foundation for monitoring the expected aerosol increase and subsequent effects. We do not want to suggest that light scattering and related optical measures will provide a sufficient basis for climatic monitoring by themselves. It is likely that a set of measurement methods used simultaneously to sense appropriate properties will emerge for some future monitoring effort.

In Section II we will define the basic physical aerosol parameters which we have measured in background locations. Section III describes the instrumentation used, and in Section IV the actual field experiments and raw data are presented. Sections V and VI discuss the data and draw conclusions.

II. Relevant optical properties of low humidity aerosols

As stated earlier, most of the data to be presented here related to low humidity, non-

cloud situations. In such cases, a relatively simple set of parameters suffices for defining the integral optical properties of particulate matter which are relevant to the problem of radiative transfer.

The extinction coefficient, b_{ext} , for a given wavelength of light is defined by the Beer Lambert law:

$$\frac{dI(x)}{I(x)} = -b_{\text{ext}}(x) dx$$

or

$$\frac{I(x)}{I(0)} = \exp \left[- \int_0^x b_{\text{ext}}(x) dx \right] \quad (1)$$

where $dI(x)$ is the decrease of intensity, $I(x)$, after passage through a length, dx , of an atmospheric path. The quantity b_{ext} is made up of two terms:

$$b_{\text{ext}} = b_{\text{scat}} + b_{\text{abs}} \quad (2)$$

where b_{scat} is the scattering component and b_{abs} is the absorption component of b_{ext} . These can be further expanded:

$$b_{\text{scat}} = b_{\text{sp}} + b_{\text{Rg}}$$

$$\text{and } b_{\text{abs}} = b_{\text{ap}} + b_{\text{ag}}$$

where b_{sp} is the scattering coefficient due to particles, b_{Rg} is the Rayleigh scattering coefficient of the gas molecules, b_{ag} is the absorption coefficient due to gases (such as O_3 , NO_2 and H_2O) and b_{ap} is the absorption coefficient due to particles. We thus have:

$$b_{\text{ext}} = b_{\text{sp}} + b_{\text{Rg}} + b_{\text{ap}} + b_{\text{ag}}$$

$$\text{or } b_{\text{ext}} = b_{\text{txp}} + b_{\text{Rg}} + b_{\text{ag}}$$

where b_{txp} is the total extinction coefficient due to particles, defined by:

$$b_{\text{txp}} = b_{\text{sp}} + b_{\text{ap}}$$

The extinction coefficient b_{ext} is perhaps the most commonly measured quantity of those listed in eq. (2). Such data are acquired as extinction coefficient integrated over altitude with pyrhelimeters, a variety of spectral solar sensors, the Volz sunphotometer and with astro-

nomical telescopes. In order to compare these total air column extinction measures with optical properties determined at a point (e.g. with light scattering instrumentation) it is useful either to measure or assume the altitude dependence of the aerosol and its properties and to correct for the absorption by ozone. In most cases, we simply assume that the aerosol extinction decreases exponentially with altitude;

$$b_{\text{ext}}(z) = b_{\text{ext}}(0) e^{-z/H_p}$$

where H_p , the aerosol scale height, is usually around 2 km. Unfortunately, this assumption has not been verified adequately. A further problem exists since high relative humidity (typically above 70%) increases the light scattering of hygroscopic aerosol (Covert et al., 1972). A layer of moist but not saturated air aloft will thus increase the extinction above the observing site even though the dry aerosol may have an exponential profile. If the high humidity air does not reach the ground, the ground level, *in situ* light scattering measurements will not be affected but the extinction data will.

Nevertheless, it is informative to compare the scattering component of extinction measured near the ground with the total extinction derived from column measurements through the atmosphere. The difference between total extinction and the scattering components might provide information on light absorption. At the very least, since it is generally agreed that $b_{\text{ap}} < b_{\text{sp}}$, the difference measured can indicate whether the assumed altitude profile is at all appropriate. The nomenclature associated with each type of measurement (astronomical extinction, turbidity, and light scattering coefficients) can be related by the following light attenuation equations:

$$I/I_0 = 10^{-km/2.5} = 10^{-(T_{\text{Rg}} + T_{\text{ozone}} + B)m} = e^{-\tau_{\text{ext}}} \quad (3)$$

where m is the number of air masses ($m = 1$ at zenith for sea level observatories), k is the astronomical extinction in magnitude per air mass, T_{Rg} is the attenuation in intensity due to scattering by air molecules per air mass, T_{ozone} is the attenuation in intensity due to absorption by ozone (for $\lambda = 500$ nm at sea level, $T_{\text{ozone}} \simeq 0.004$ (Flowers et al., 1969), B is the decadic turbidity coefficient and τ_{ext} is the total extinction of the atmosphere, often called optical

depth. These quantities can be related to those defined earlier by:

$$T_{\text{Rg}} = \frac{1}{2.3} \int_0^\infty b_{\text{Rg}}(z) dz = \frac{1}{2.3} b_{\text{Rg}}(0) H_{\text{Rg}}$$

where z is the vertical coordinate, H_{Rg} is the molecular scale height (~ 8 km at sea level) $b_{\text{Rg}}(0)$ is the value of b_{Rg} at $z = 0$ (sea level), and:

$$B = \frac{1}{2.3} \int_0^\infty b_{\text{txp}}(z) dz$$

so if $b_{\text{txp}}(z) = b_{\text{txp}}(0) e^{-z/H_p}$, then

$$B = \frac{1}{2.3} b_{\text{txp}}(0) H_p$$

where $b_{\text{txp}}(0)$ is the value of b_{txp} at $z = 0$. These quantities must be corrected for altitude when observations are made at $z \neq 0$.

The scattering coefficient due to aerosol, b_{sp} , is itself an angular integral from 0 to π . Only part of this integral contributes to the albedo since light scattered downward is in a first approximation the same as transmitted light. In the simplest case of a flat earth and zenith sun, the hemispheric backscattering coefficient, b_{bsp} (which is the part of the extinction coefficient due to scattering by particles in the angular range from $\pi/2$ to π).

$$b_{\text{bsp}} = b_{\text{sp}} - b_{\text{fsp}}$$

where b_{fsp} is the scattering coefficient due to particles for angles from 0 to $\pi/2$ and results (in the zenith sun case) in light reaching the ground essentially along with the transmitted light.

It is important to recognize that the fraction $b_{\text{bsp}}/b_{\text{sp}}$ is not a well known quantity and that it depends on both size distribution and particle shape (Harrison et al., 1972; Waggoner et al., 1972). Several workers (Atwater, 1970; Mitchell, 1971; Ensor et al., 1971) have shown that for simple model atmospheres, the relationship of b_{bsp} to the aerosol absorption coefficient (b_{ap}) is needed if the climatological effect of aerosol is to be assessed. Hence it is useful to obtain measurements of the hemispheric backscatter which may ultimately be compared to b_{ap} . While we do not herein compare b_{bsp} and b_{ap} , the method to be presented makes the comparison possible.

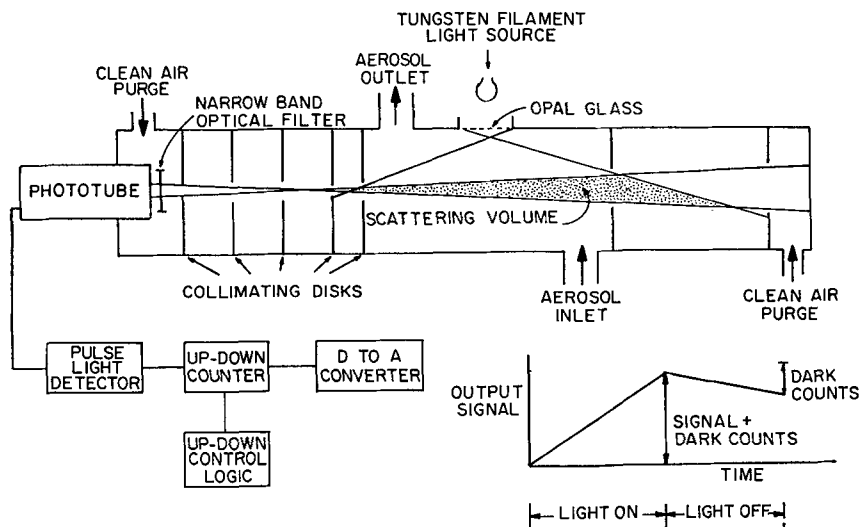


Fig. 1. Photon counting modification to the integrating nephelometer.

III. Instrumentation

The basic design of the integrating nephelometer used in these experiments was derived from Beuttel & Brewer (1949) and from Crosby & Koerber (1963), and has been published previously (Ahlquist & Charlson, 1968). Briefly, integrating nephelometers integrate the volume scattering function ($\beta'(\phi)$) over a range of scattering angles (from $\phi \approx 0$ to $\phi \approx \pi$). The inclusion of a sine weighting function in the integrand, accomplished by the use of a Lambert (cosine) source normal to the scattering axis results in a close approximation to the integrand controlling the scattering component of the extinction at the location of the integrating nephelometer:

$$b_{\text{scat}} = 2\pi \int_0^\pi \beta'(\phi) \sin \phi d\phi \approx \int_{7^\circ}^{170^\circ} \beta'(\phi) \sin \phi d\phi$$

where $\beta'(\phi)$ is the volume scattering phase function defined by Middleton (1952) at angle ϕ from the transmitted beam. The accuracy of this integration over the real instrumental range from 7° to 170° will be discussed later.

A. Photon counting adaptation

It was necessary to modify the existing nephelometer for (1) higher sensitivity to b_{sp} in high altitude background locations and (2)

measurement of the albedo contribution of aerosol scatter, b_{bsp} .

Higher sensitivity was achieved by changing the light source and method used to detect the scattered optical signal. In the earlier nephelometer a xenon flash lamp produced ~ 10 microsecond pulses of light and the signal from a photomultiplier tube was detected with peak reading amplifiers. In the new technique, the light source is a tungsten filament incandescent lamp and the scattered light is detected by digital photon counting methods.

The light source, nephelometer, and a box diagram of the signal electronics are shown in Fig. 1. The signal from the photomultiplier tube consists of pulses due to electrons emitted from the photocathode, pulses from electrons thermally emitted from points within the dynode chain, and leakage current between elements within the tube. Only electrons from the photocathode are useful signals; the other currents are noise. The pulses due to photoelectrons are larger because they have been amplified by the entire dynode structure and can be detected by a level discriminator that produces a standard output pulse for each electron emitted by the photocathode.

The detected pulse rate would be proportional to the light intensity plus a temperature-dependent dark current due to thermal emission of electrons from the photocathode. (In our

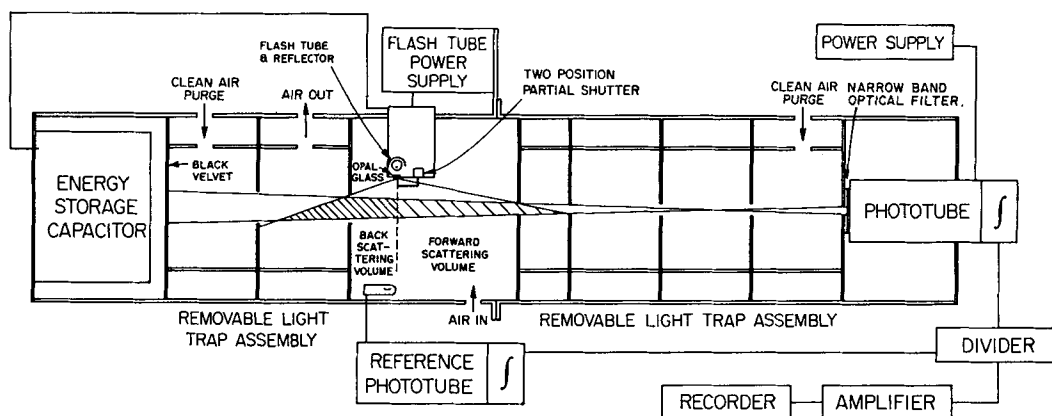


Fig. 2. Integrating backscatter-total scatter nephelometer.

experiment the dark current count rate was about three hundred per second and the Rayleigh scatter signal was about three times higher.) The thermal emission signal is removed by using reversible electronic counters.

A data cycle consists of four stages:

1. Lamp is on at regulated intensity and the counter is accumulating counts upward from zero.

2. After a regulated time interval the counter is put into hold and the lamp is turned off.

3. The counter is put into count down status and allowed to count for a time interval equal to the light on period.

4. The counter is put into hold status and the digital count is converted to a proportional analog signal and recorded. The counter is reset to zero, the light turned on, and step 1 takes place again.

This logic and operational sequence subtracts the counts due to thermal emissions from the signal measured during the light-on interval. This form of data handling has very low drift, attains the highest possible accuracy for a given light signal and has achieved an improvement in sensitivity of a factor of ten to twenty over our best previous nephelometer.

B. Hemispheric backscatter modification

The nephelometer internal optics have been modified to allow measurement of aerosol scatter integrated over two different angular ranges. A two position partial shutter, shown in Fig. 2 can be placed so that the integrated backscattering and total scattering component of ex-

tinction can be determined by changing the position of the partial shutter. This allowed two ranges of acceptance angles: from 8° to 170° and 90° to 170° . The operating procedure for the instrument is similar to that for past instruments. The instrument backscatter/total scatter ratio is calibrated by measuring the ratio for clean air, He, CO_2 and/or Freon-12. The measured ratio of backscatter to total scatter ($b_{\text{bsg}}/b_{\text{Rg}}$) for gases as expected from the geometry of the instrument and the symmetry of the phase function for Rayleigh scattering was 0.50 ± 0.04 . The general description of the use of gases for calibration is included below.

C. Calibration and accuracy

As described by Charlson & Ahlquist (1969) the calibration of the nephelometers is routinely accomplished with absolute-filtered standard gases. The Rayleigh scattering of air is used as the primary standard, with CO_2 , Freon 22, and Freon 12 providing upscale calibration points and helium providing a value near zero. Bhardwaja et al. (1973) reported the measured ratios of the scattering coefficient for these gases relative to that of air:

Gas	b_{Rg} (calibration gas)/ b_{Rg} (air)
He	0.01 (calculated)
CO_2	2.61 ± 0.03
Freon 22 (CHClF_2)	7.72 ± 0.09
Freon 12 (CCl_2F_2)	15.62 ± 0.18

These gases are taken from compressed gas or liquid containers to insure purity and absence of aerosol and subsequently are filtered through an absolute filter of guaranteed $99 \pm \%$ efficiency prior to use in a calibration.

Other means may be used for this calibration, including geometric optics schemes such as that described by Middleton (1952) or more recently by Horvath (1973). The chief advantage of using Rayleigh scattering gases is their nearly perfect reproducibility as standards (if properly filtered). Mechanical/optical devices such as diffuse reflectors or opal glass require accurate positioning and must have constant reflectivity or transmission properties over time. It is difficult to match or maintain the reflection or transmission properties of such materials within a few percent, and dirt or deposited aerosol on their surfaces cause errors. Most importantly the gases permit evaluation of the amount of scattering from the apparatus itself.

Thus particle free Rayleigh scattering gases provide unequivocally reproducible standards for nephelometer calibration. This aspect of these measurements is critically important since we wish to study secular trends over extended periods of time—decades or more. In such cases, standard scattering gases should provide the necessary confidence in calibrations for detection of even small trends.

It is necessary to define the accuracy, precision and detection limit of the measured values of b_{sp} for atmospheric aerosol. The main source of systematic errors governing the accuracy is the truncation of light scattering at angles between 0° and 7° and from 170° to 180° . Middleton (1952), Ensor & Waggoner (1970), Rabinoff & Herman (1973) and Heintzenberg & Quenzel (1973) have calculated the error caused by this truncation for a wide variety of particle size distributions. The general conclusion is that the typical error in the absence of large dust particles or fog is from -2 to about -10% . This error is systematic in that it is characteristic of a given size distribution and refractive index.

The precision or reproducibility of the nephelometer measurements is determined by the signal/noise ratio and by drift between calibrations. The precision without using any differential measurement techniques is ca. $\pm 5\%$ of b_{scat} . Using a differential technique, this figure can be improved and also permits the lowering of the detection limit.

D. Differential technique

By switching the sample air flow intermittently (say, ca. every 30 minutes) from ambient to absolute filtered air, a differential measurement can be made which obviates the small amount of random drift in instrument zero or changes in atmospheric density due to temperature or pressure. By ascertaining that the instrument sensitivity was in fact constant within $\pm 2\%$, differential technique permitted the measurement of values of b_{sp} which were less than 0.5% of b_{Bg} under field conditions at the Mauna Loa Observatory at a wavelength of ~ 500 nm.

Due to sensitivity requirements in background locations, it was often necessary in the earlier work to use broad wavelength bands, with a typical width of ca. ± 50 nm. In the earliest instruments (Porch et al., 1970) the wavelength definition was provided by a UV cutoff filter (Wratten 2A) and an S-11 phototube response. Data were subsequently corrected from this very broad band to a wavelength of 550 nm by use of estimates of the wavelength dependence of aerosol scattering. The photon counting and backscatter total scatter device utilized a Wratten 58 filter combined with an S-20 photocathode to yield a band at 525 ± 25 nm which is sufficiently monochromatic to greatly reduce the need for correction of wavelength. Much finer wavelength definition has also been used in a multiwavelength integrating nephelometer (Ahlquist & Charlson, 1969), using 5 to 30 nm bandpass filters. Data from this device have been correlated with the broader band instruments yielding correlation coefficients, r , in a range from $0.94 \leq r \leq 0.99$ in urban aerosols.

IV. Description of field experiments

A. Mauna Loa Observatory

The high sensitivity nephelometer was located at the National Oceanographic and Atmospheric Administration Observatory at 3 350 m altitude on Mauna Loa and at the high point of the Mauna Loa–Mauna Kea saddle (2 134 m). The backscatter/total scatter nephelometer was located at Cape Kumukahi near sea level. The long-term data on CO_2 content at the Mauna Loa Observatory (MLO) presented by Machta (1972) provide an excellent baseline measurement of this atmospheric constituent. Analogous meas-

Table 1. *Light scattering data at 500 nm*

Site, State (altitude in meters)	Dates	$\frac{b_{\text{scat}}}{b_{\text{Rg}}}$	$\frac{b_{\text{bsp}}}{b_{\text{sp}}}$	Comments
Mauna Loa Observatory, HI (3370)	5-8 March 1971	$1.07^{+0.05}_{-0.04}$	—	Preliminary experiment to determine instrument requirements. b_{scat} during downslope winds including 90 % interval
Mauna Loa Observatory, HI (3370)	21 Feb.- 14 Mar. 1972	$1.10^{+0.10}_{-0.07}$	—	Geometric mean of 123 hourly averages using differential techniques, subsiding air only
Cape Kumukahi, HI (~10)	17 Feb. 20 Mar. 1972	$3.8^{+0.7}_{-0.6}$	$0.13^{+0.03}_{-0.02}$	Land breeze, 62, 15 minute averages
Cape Kumukahi, HI (~10)	17 Feb. 20 Mar. 1972	$2.6^{+0.6}_{-0.9}$	0.1 to 0.2	Sea breeze
North Pacific Ocean, ship (15)	1 Sep.- 10 Oct. 1971	$2.3^{+0.65}_{-0.35}$	—	Geometric mean of 462, 30 minute averages, including 90 % interval
Mount Olympus, WA (2025)	12 Feb.- 28 Mar. 1968	$1.5^{+0.30}_{-0.25}$	—	Geometric mean of 1 400 hourly averages including 90 % interval
Manastash Ridge, WA (1, 141)	26 Oct. 8 Nov. 1971	$2.3^{+2.35}_{-1.22}$	—	Geometric mean of 304, 30 minute average with 90 % interval
Point Reyes, CA (190)	16 Aug.- 25 Aug. 1972	$1.24^{+4.30}_{-0.37}$	—	Geometric mean of 770, 10 minute averages with 90 % interval
Point Reyes, CA (190)	23 Aug.- 25 Aug. 1972	$3.76^{+4.61}_{-3.00}$	0.06	Geometric mean of 197, 10 minute averages with 90 % interval. This $b_{\text{bsp}}/b_{\text{sp}}$ ratio results from only 20, 10 minute averages and no 90 % is provided. Wind to 20 ms ⁻¹

urements of turbidity at the Mauna Loa Observatory to 1972 indicate an absence of increases in aerosol extinction due to human activity up to now, making it an ideal location for background aerosol studies (Machta, 1972).

Measurements of b_{sp} were made with the photon counting device at MLO from 21 February to 14 March 1972. The instrument was located in a shelter, with a 3 cm diameter intake at a height of 2 m above ground. Data were recorded continuously on a Leeds and Northrup model W recorder and hand reduced to tractable form.

Another preliminary experiment at MLO was conducted from 5 to 8 March 1971 to determine the sensitivity requirements for the instrument used in 1972. The data from both periods are included in Table 1 and are presented graphical-

ly in Fig. 3. These data are discussed in the next section.

As in the CO₂ studies at MLO, marked differences were found in all data during upslope and downslope wind conditions. Only the data during subsiding or downslope conditions qualify as having baseline character, the upslope data being affected by the aerosol from the island itself or from the surrounding ocean. Care was taken to make sure that locally generated dust had not influenced the measurements by examining the record for the short term (1-15 minute) fluctuations characteristic of dust generated by turbulence. While upslope data occasionally showed such evidence of recently-generated puffs or clouds of dust, the downslope data exhibited only slow variations. This results in a smaller geometric standard deviation.

tion in the case of downslope winds as illustrated in Fig. 3.

B. North Pacific Ocean

The experiment was performed with an earlier version of the nephelometer on the oceanographic research vessel Thomas G. Thompson as part of a major oceanographic project comparing sediment transport by turbidity currents in the deep ocean and sedimentation by aeolian transport. For this purpose, the ship was stationed for several days over the deep ocean Nitinat Fan ~240 km from the coast. From Nitinat Fan it navigated south to ~64 km west of the Columbia River then north to ~160 km from the northern tip of Vancouver Island.

The period of record extends from 1 September to 10 October 1971, during which time the prevailing synoptic situation provided air which had a recent maritime rather than continental history. The instrument was located in the top deck navigation cabin, with the intake extend-

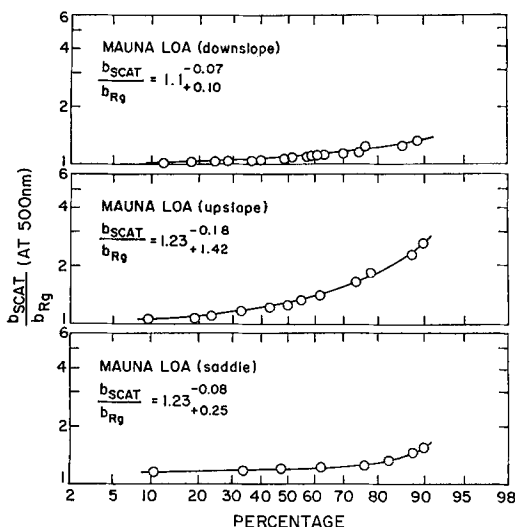


Fig. 3. Logarithmic probability plot of the ratio of observed scattering (b_{scat}) to Rayleigh scattering by air (b_{Rg}) at 500 nm at Mauna Loa Observatory, Hawaii (3 370 m) for downslope winds or subsiding air with 123 hourly averages, for upslope winds with 42 hourly averages and at the Mauna Loa Saddle (2 134 m) with 59 fifteen minute averages from 17 February to 20 March 1972. Shown on the ordinate is the ratio; values along the abscissa represent the percentage less than the ordinate value. In this plot the ordinate value corresponding to the abscissa value of 50 % is the geometric mean, and the slope gives the geometric standard deviation.

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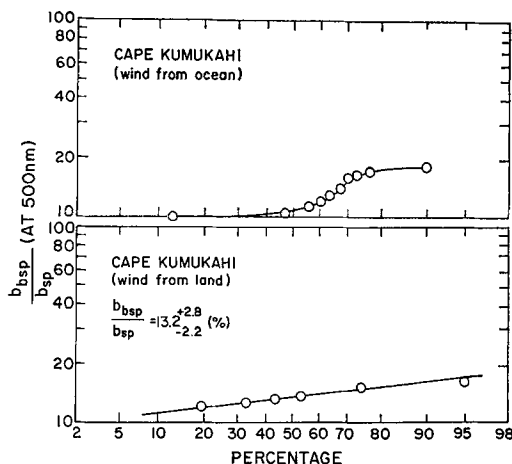


Fig. 4. Logarithmic probability plot of the ratio of observed aerosol backscatter (b_{bs} integrated scatter from ~90° to ~180°) to aerosol total scatter (b_{ts} integrated scatter from ~0° to ~180°) at 500 nm at Cape Kumukahi Hawaii (~sea level) for winds originating from the ocean with 70 fifteen minute averages and for winds originating from the land with 62 fifteen minute averages from 17 February to 20 March 1972.

ing ca. 10 meters up and projecting in front of the ship's mast. The angle from the intake to the ship's bows was ca. 30 degrees, providing a clean air stream not perturbed by the dust from the decks or by spray from the bow wave. Data included in Table 1 were taken only when steaming to windward. The sea state varied considerably during this period; however no correlation between the scattering properties and sea state was immediately obvious.

C. Mt. Olympus, Washington

These measurements were made with an earlier broadband version of the integrating nephelometer. The station on Mount Olympus is located on a precipice on the northwest side of this mountain, which is located on the sparsely populated Olympic Peninsula. This area is usually upwind of virtually all urban and industrial aerosol sources in Washington and British Columbia due to its exposure to prevailing westerly air flow from the Pacific Ocean.

The data from this experiment were reported earlier by Poreh et al. (1970) and were taken between 12 February and 28 March 1968.

D. Point Barrow, Alaska

These measurements were made with an ear-

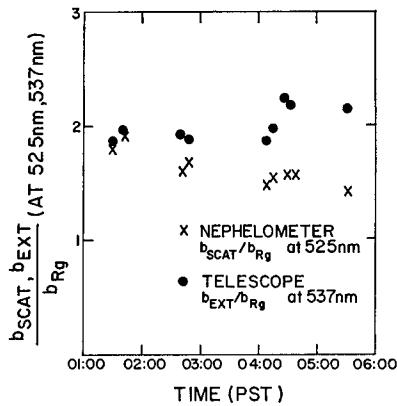


Fig. 5. Total scattering coefficient divided by molecular scattering coefficient ($b_{\text{scat}}/b_{\text{Rg}}$) as determined by the nephelometer at 525 nm and the analogous ratio determined by astronomical techniques ($b_{\text{ext}}/b_{\text{Rg}}$) at 537 nm during a night of simultaneous observations with assumed optical thickness of ozone of 0.35 cm.

lier version of the integrating nephelometer. The nephelometer was installed in a remote micro-meteorological hut, located 5 km north-east and upwind of the village of Point Barrow. The Beaufort Sea and the Arctic Ocean were covered with ice ~3 m thick making locally derived sea salt aerosol a negligible component of the aerosol. The period of record was 10 to 25 March 1970.

E. Manastash Ridge, Washington

Nephelometer and astronomical extinction measurements were made briefly from 26 October to 8 November 1971 at the University of Washington Observatory on Manastash Ridge, located 16 km SW of Ellensburg, Washington. This site is not expected to often be baseline in character due to the cities to the west (Vancouver, B.C., Seattle and Portland) from which direction the prevailing winds may be expected to transport aerosol. Nonetheless, under selected meteorological conditions, the air here should be dominated by natural aerosol. The chosen period was post-frontal and cloudless with recent deep convective mixing. The rationale in this selection was to provide a uniform air mass above the observatory so that local extinction (via the nephelometer) could be compared to the astronomical extinction. During the period of the experiment, clear nights were rare and simultaneous astronomical extinction and neph-

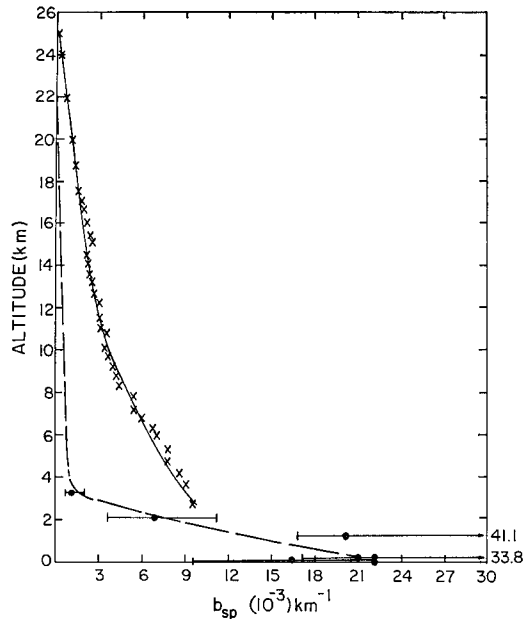


Fig. 6. Altitude dependence of light extinction due to aerosols by nephelometer and astronomical telescope, with searchlight data of Elterman (1973) for comparison.

Fig. 6a. Nephelometer data with 90 % interval for b_{sp} from Table 1 at 500 nm (---) compared to searchlight data (—) at 550 nm of Elterman (1973).

elometer data were possibly only on one night during this period (November 5). A comparison of data for this night is given in Fig. 5.

F. Other Astronomical Extinction Comparisons

A large quantity of astronomical extinction data have been acquired by several groups in recent years (Hodge et al., 1972) against which the nephelometric data may be compared. A few of these data are included here to illustrate the basic nature of such measurements. Eventually, it is anticipated that a large quantity of such telescopic data, both from past decades and from current observations, will be available for aerosol studies. Fig. 6 includes data from 17 observatories, all of which are located away from human activity.

G. Point Reyes National Seashore, California

The nephelometer was located in a laboratory van parked at the US Coast Guard Station at Pt. Reyes, 60 km northwest of San Francisco.

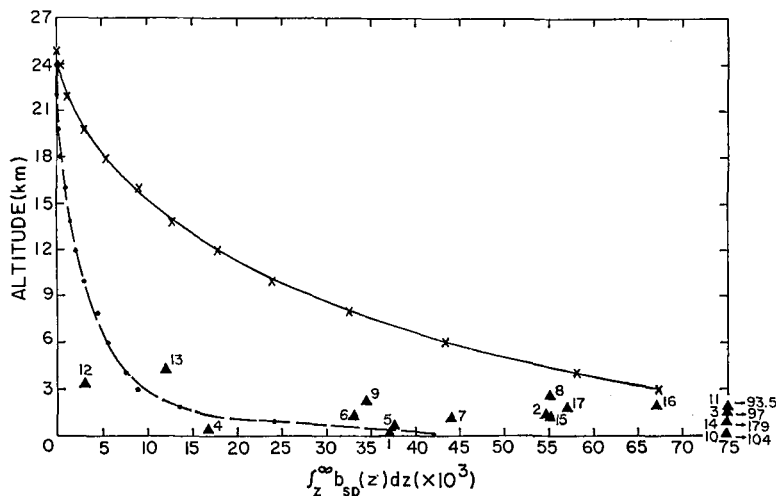


Fig. 6b. Mean aerosol optical depth versus altitude via astronomical telescope (data points) at 500 nm, the integral $\int_0^\infty b_{sp}(z)dz$ for the geometric mean nephelometer data in Fig. 6a (---) and the same integral for the searchlight data (—). The astronomical data compared here can be regarded as typical for remote locations, and were taken over varying time periods by many different observers. Details of the sources of these data, dates and techniques will be presented in a forthcoming paper. A value of $T_{\text{ozone}} = 0.004$ was assumed in the telescopic data. Observatories are by number, with altitude (meters) in parentheses: (1) Royal, Cape of Good Hope (10), (2) Boyden, Republic of South Africa (1387), (3) Radcliff, Republic of South Africa (1542), (4) Mt. Bingor, Australia (460), (5) Mt. Stromlo, Australia (768), (6) Siding Spring, Australia (1164), (7) Mt. John, New Zealand (1029), (8) Europ South, Chile (2400), (9) Cerro Tololo, Chile (2195), (10) Le Houga, France (146), (11) Naini Tal, India (1927), (12) Mauna Loa, Hawaii (3370), (13) Mauna Kea, Hawaii (4170), (14) Rattlesnake Ridge, Washington (1080), (15) Lick, California (1283), (16) Kitt Peak, Arizona (2064), (17) McDonald, Texas (2081).

Situated ca. 190 m above the sea on a cliff, this site is exposed to onshore flow from southerly through westerly to northerly directions. A lack of public access minimize locally generated particles. Frequent episodes of extremely dense fog, however, made aerosol sampling extremely difficult. Droplets including hydrated aerosol were efficiently deposited in the sampling intake, providing essentially no aerosol to the instruments. Data reported in Table 1 include periods selected only when fog was absent. Substantial parts of the period of record were taken under high wind conditions (15 to 20 ms^{-1}) with strong onshore flow providing another marine background site. One 20 hour period of higher winds from the NW had a nearly constant b_{sp} , as reported in Table 1.

V. Data discussion

The nephelometer data from these remote sites are shown in Table 1, with 90% of the data points falling between the upper/lower limit in-

dicated. The data for b_{scat} are reported as the ratio to Rayleigh scattering, i.e. b_{scat}/b_{Rg} , and are all for a wavelength of 500 nm.

1. Except for the data taken at Manastash Ridge (under selected weather conditions), scattering and extinction levels show little variation (i.e., 90% interval is small) during the measurement periods at the various locations. This is expected if the levels of atmospheric aerosol at these places is not influenced by local sources or sinks.

2. There appears to be a detectable difference in the backscattering properties of atmospheric aerosol from one location to another.

3. As expected, total aerosol light extinction exceeded total aerosol light scatter at the locations where the nephelometer and telescope techniques were applied simultaneously. However, until better resolution of the aerosol distribution with height can be made (perhaps with lidar or airplane techniques) reliable estimates of b_{ap} cannot be made from the difference. It is apparent from the near equality of b_{scat} and b_{ext} that during the experimental

period the stratospheric aerosol made little contribution to total extinction. This is not the case during periods of high volcanic activity.

4. Even though the background scattering and extinction ratios shown in Table 1 vary with location, altitude, and the period of time during which the observations were made, the background levels seem to depend more on altitude than location or time. The altitude dependence itself exhibits some regularity from site to site. The altitude dependence of the geometric means are compared to the data of Elterman (1973) in Fig. 6a and b.

The results of the MLO experiment are shown in Figs. 3 and 4 as logarithmic probability graphs of the light scattering data. On the ordinate are the ratios $b_{\text{scat}}/b_{\text{Rg}}$ (where $b_{\text{scat}} = b_{\text{sp}} + b_{\text{Rg}}$) and $b_{\text{bsp}}/b_{\text{sp}}$. The values along the abscissa represent the percentage less than the ordinate value. In this plot the ordinate value corresponding to the abscissa value of 50% is the geometric mean, and the slope gives the geometric standard deviation. Fig. 3 compares the ratio of $b_{\text{scat}}/b_{\text{Rg}}$ at 500 nm wavelength at the Mauna Loa Observatory for downslope (subsiding upper tropospheric air) and upslope (rising air) winds of less than 25 mph and data taken in the saddle below the observatory. Under higher wind conditions the light scatter increased appreciably at times indicating the possibility of contamination by local wind blown surface dust. Under ordinary circumstances the data trace showed little short-term fluctuation due to small scale eddy transport of local dust. The ratio $b_{\text{scat}}/b_{\text{Rg}}$ measured at the observatory with 123 one hour averages represents the lowest levels of b_{sp} and the most consistent light scattering data ever taken with our instrument. The geometric mean ratio of $1.1_{-0.10}^{+0.07}$ compares to $1.5_{-0.25}^{+0.30}$ for 1400 hourly averages at Mount Olympus, Washington and $1.95_{-0.40}^{+0.35}$ for 650 hourly average at Pt. Barrow, Alaska (Porch et al., 1970). The $b_{\text{scat}}/b_{\text{Rg}}$ ratio shown in Fig. 3 under upslope wind conditions represents 42 one hour averages. The varying slope seems to indicate the effect of two types of air involved under upward transport conditions:

1. The relatively clean air under downslope wind conditions associated with little aerosol scattering.
2. The air associated with the saddle region

below the observatory with a relatively higher level of light scattering.

The $b_{\text{scat}}/b_{\text{Rg}}$ ratio shown in Fig. 3 for the saddle region (1216 m below the observatory) represents 59 fifteen minute averages made mostly at night. It is interesting to note that the geometric mean value of $1.23_{-0.25}^{+0.08}$ is the same as that measured at the observatory under upslope conditions. This substantiates the close association of the air masses under these conditions.

It is interesting to compare the magnitude of b_{sp} at the altitude of MLO with that obtained by Elterman (1973) (see Fig. 6). It is apparent that either there is less aerosol at MLO and at our other sites than in New Mexico, or that there is a systematic error in either or both sets of measurements. One possibility is the necessity of assuming a phase function in interpreting searchlight data such as those of Elterman. Another possibility is that searchlight data can only be acquired on clear, cloudless, moonless nights which biases these data to given synoptic cases and thus to selected aerosol histories.

Shown in Fig. 4 are logarithmic probability plots of the $b_{\text{bsp}}/b_{\text{sp}}$ ratio at 500 nm measured with the integrating backscatter/total scatter nephelometer at Cape Kumukahi under wind conditions from and away from the ocean. The lack of continuity of the slope of the ratio for winds from the ocean seems to indicate the effect of varying amounts of surf spray aerosol. Since these particles are ordinarily quite large their effect is to diminish the ratio of $b_{\text{bsp}}/b_{\text{sp}}$ when they are present.

Filter samples were taken during part of the measurement period at the Mauna Loa Observatory during one 18 hour interval and one 7 hour interval. The mass concentration (M) to light scatter ratios (M/b_{sp}) proved to be $5.5 \times 10^5 \mu\text{g}/\text{m}^2$ and $3.3 \times 10^5 \mu\text{g}/\text{m}^2$ respectively. These values compare quite closely to the mass to light scatter ratio determined by Charlson (1969) for urban locations of $4.5_{-2.2}^{+4.5} \times 10^5 \mu\text{g}/\text{m}^2$.

Besides the data on light scattering levels in background sites, a small but growing body of data is being developed regarding the wavelength dependence of scattering between 300 and 800 nm. Porch et al. (1971) reported wavelength dependences of astronomical extinction measurements, and Porch et al. (1973) analyzed these in terms of aerosol size distributions. In all of these cases, a substantial fraction (perhaps

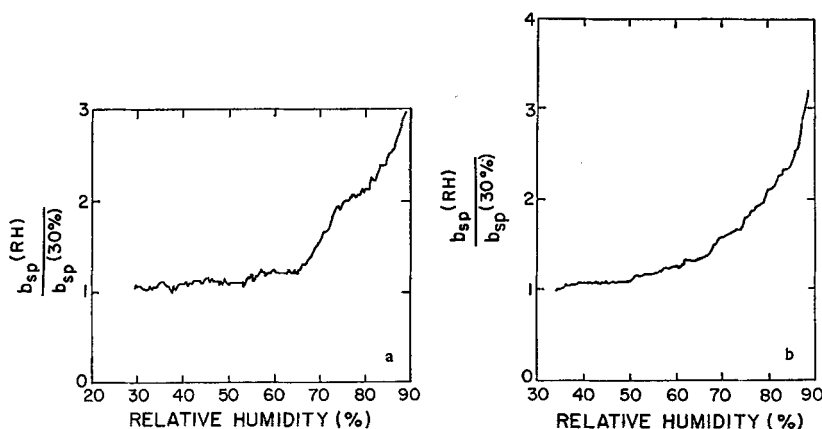


Fig. 7. Light scattering, b_{sp} , normalized to that at 30 % humidity versus relative humidity at Pt. Reyes, California showing the presence of sea salt aerosol (a) and a non salt aerosol (b) for comparisons.

50 %) of the data showed an increase of light scattering in the red. If the approximation that

$$b_{sp} \propto \lambda^{-\alpha}$$

is used, then α was often negative, quite in contrast to the situation in urban aerosol described by Charlson (1972).

Several measurements at low altitude in marine air, particularly at Pt. Reyes, California, show this same feature with $-0.5 < \alpha < 0.5$. While it is too soon to claim a general result based on few data, this feature may well be a regular characteristic of well aged aerosol.

The effect of relative humidity on light scattering properties is also under study by the method of Covert et al. (1972). The rationale for this study is the fact that the lower troposphere often has relative humidities (RH) in the range $30 \leq RH \leq 90\%$ or higher, and that the role of condensed water due to hygroscopic and/or deliquescent substances may not be neglected. Since on a short time scale the dry aerosol mixing ratio may be assumed to be conserved, the importance of humidity is to always increase the observed scattering levels for a given dry aerosol concentration.

Preliminary data in remote marine locations clearly show the presence of NaCl aerosol as a deliquescent salt. An example of such a curve is shown in Fig. 7, in which a non-salt aerosol in the same location is included for comparison. These observations show conclusively that

humidity in the air column above a turbidity or astronomical extinction observer must be accounted for.

VI. Conclusions

The preliminary data collected by means of nephelometers and reported in this paper are consistent with other optical measures (turbidity and astronomical extinction) of background aerosol at various heights particularly above 2 km but are not consistent with searchlight data. The amount of data collected is large in spite of the relatively short sampling periods. We find that in background locations $b_{scat}/b_{Rg} \approx 1.1-2.3$ and $b_{bsp}/b_{sp} \sim 10-20\%$, with anomalous wavelength dependence ($b_{sp} \sim \lambda^{-\alpha}$, $\alpha < 0$) approximately 50 % of the time.

The chief advantages of the nephelometer for background aerosol surveillance are its automatic and continuous operation and reproducibility by use of gases for calibration. Turbidity and astronomical extinction measurements require an observer so that the number of data points acquired in a given time period is very small compared to the number collected by a continuous instrument. Turbidity and extinction measurements also are confined to cloudless weather, and can be performed only at low air mass number during day and night respectively. The necessity of cloudless weather biases these aerosol measurements since they selectively include only certain synoptic situations and hence

they exclude situations with their peculiar air mass and aerosol histories. Rarely is a Lidar available to detect thin clouds that are not visible to the observer.

Inspection of the slopes of the frequency of occurrence graphs (Fig. 3) further confirms the hypothesis that the geometric standard deviation (i.e. the slope) is a useful index of baseline character for a given site. Sites where local sources were apparent had larger or undefinable values. It can be inferred from these observations that light scattering data are useful in site selections and that in conjunction with condensation nuclei measurements they provide information that is useful in defining aerosol system parameters.

All the optical measurements made at a given wavelength sense the same range of particle sizes, usually from $0.1\ \mu\text{m}$ to about $1.0\ \mu\text{m}$ in diameter. Exceptions occur in fog or dust and sand storms, where the optical effects are due to particles of up to perhaps $\sim 10\ \mu\text{m}$ diameter. In contrast, condensation nuclei counters, which have been used for study of secular trends of aerosol sense much smaller particles with a range of perhaps $10^{-3}\ \mu\text{m} \leq \text{diameter} \leq 10^{-1}\ \mu\text{m}$. Hence, these different methods measure different and probably quite independent aerosol system properties. It thus seems necessary to utilize these and other methods simultaneously as suggested by Jaenicke (1973) to adequately monitor aerosol. In light of this realization, it is obviously hazardous to use condensation nuclei count alone to determine whether a site is suitable for baseline measurements of aerosol. Rather, such exploratory measurements must consider the existence of the other relevant size classes. One-parameter characterizations of aerosol at best provide an incomplete picture and are often found to be misleading when aerosol system parameters are needed.

Even though the light scattering measurements presented above must be used with other measurements in order to characterize the aerosol system, they do provide relevant quantities that can be used in a variety of optical models of the atmosphere. In view of the complexity of the aerosol system and the difficulty of calculating the relevant parameters from a few bits of data, it is particularly desirable to seek measurements whenever possible that are in themselves useful and relevant. For example

the hemispheric backscatter (b_{bsp}) is a property of aerosol that can be used, for instance in radiative balance models, while the condensation nuclei count is not known to be related to meteorological factors in any simple way. Of course, no conclusions may be drawn regarding secular trends; sampling periods of the order of decades will be required when such monitoring is established.

The key answers to questions regarding the climatological effects of aerosol remain as somewhat closer but still unattained goals. The absorption coefficient, which can be estimated by the method of Lin et al. (1973) must be determined simultaneously with the hemispheric backscatter if the heating versus cooling controversy is to be resolved.

Even then, the problem remains of the effect of distribution of the aerosol (Charlson et al., 1972) both horizontally and vertically. Most of the aerosol extinction is below 3 km of altitude even in remote locations. Many clouds are at or below this height as well, so that cloud/aerosol interaction is mostly below 3 km. Hence, we conclude that the main meteorological effects of aerosol (natural or anthropogenic) are likely to occur at low altitude, except where there is direct injection of aerosol at higher altitudes, i.e. rockets, SST aircraft, or massive volcanic eruptions.

It is true as Elterman (1973) suggests that stratospheric aerosol is long lived and therefore more nearly global in extent while tropospheric aerosols are limited except in rare cases to smaller regions near their sources. However, we take issue with the implication that tropospheric aerosol is unimportant for global climate. Stratospheric aerosol effects on global climate will no doubt be easier to explain, in part because of the lack of high humidity hazes and clouds in the stratosphere and the much simpler radiative models that have been developed. Even though the troposphere situation is more difficult to monitor and to model due in part to spatial and temporal variability, secular trends in weather and climate on a global scale due to increased aerosol in the lower atmosphere certainly cannot be dismissed. Of special importance in the case of tropospheric aerosols is the possibility for systematic changes in synoptic patterns due to either the radiative or nucleation effects of high concentrations of aerosol systematically located in a few regions

of the northern hemisphere. Also, aerosol-induced changes in vertical convective motion in the lower atmosphere and/or aerosol-induced changes in lapse rate may occur (Schneider, 1972).

For this very reason, it may be much more important to monitor the tropospheric aerosol within one or two removal distances of its source regions (e.g. the continent of Europe, the eastern US, etc.) rather than exclusively at remote sites like MLO or in the stratosphere. We believe that a) more measurements relevant to climate of both stratospheric and tropospheric aerosols are needed and b) that better models of the effects of aerosol on weather and climate using such measurements are also needed before it will be possible to state unequivocally whether stratospheric or tropospheric aerosol dominates. For that matter,

more measurements and better models are needed in order to state to what degree aerosol is important to each of the several effects.

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ОСНОВНЫЕ СВЕТОРАСSEИВАЮЩИЕ ХАРАКТЕРИСТИКИ ФОНОВОГО АЭРОЗОЛЯ: НАБЛЮДЕНИЯ ЛАБОРАТОРИИ МАУНА ЛОА В СРАВНЕНИИ С РЕЗУЛЬТАТАМИ ИЗМЕРЕНИЙ В ДРУГИХ УДАЛЕННЫХ ПУНКТАХ

Фоновый аэрозоль может быть определен как существующий в удаленных пунктах, достаточно далеко от локальных переменных источников. Общий вклад в коэффициент экстинкции оптического аэрозоля из-за рассеяние в заднюю полусферу важны из-за их роли в определении атмосферной экстинкции и альбедо соответственно. Описывается специальная аппаратура для измерения этих параметров в удаленных пунктах на различных высотах в тропосфере. Описаны результаты предварительных измерений и сравниваются данные для нескольких пунктов и высот. Обсерватория Мауна Лоа (Гавайи)

представляет особый интерес, т. к. она используется для изучения увеличения содержания CO_2 . Измеренный коэффициент рассеяния света, обусловленный аэрозолем при длине волны 500 мкм составляет 10 % от релеевского рассеяния на газовых молекулах и указывает направление для будущих изменений. Результаты полусферического обратного рассеяния (или альбедо) указывают на величину, большую, чем коэффициент, предсказанный формулой Ми, и тем самым говорят о распределении сферических частиц по размерам.