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AEROSOL OPTICAL PROPERTIES OF THE FREE TROPOSPHERE

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In two balloon flights over Laramie, Wyoming, simultaneous measurements of aerosol backscatter and total aerosol scatter were successfully obtained. The data have been analyzed and used to directly measure the extinction-to-backscatter ratio under the assumption that the aerosols are relatively non-absorbing (small imaginary component to the index of refraction). The results are consistent with previous estimates of the extinction-to-backscatter ratio, although very little information is available relevant to direct measurements of this important parameter. It is concluded that the new instruments and techniques employed here will provide a practical method of obtaining a wealth of new information concerning the extinction-to-backscatter ratio of tropospheric aerosols.

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SUMMARY

This year's effort was centered around a unique and direct measurement of the extinction-to-backscatter ratio for tropospheric aerosols in the boundary layer and free troposphere. The work involved extensive calibrations of the ground based integrating nephelometer standard and two balloon flights in which simultaneous measurements of backscatter and total aerosol scattering were obtained.

1. INTRODUCTION

The ratio of extinction-to-backscatter for aerosols throughout the troposphere is an essential parameter for understanding radiative transfer properties and utilizing information obtained from some types of optical remote sensing devices. Since tropospheric aerosol particles in general may have irregular shapes and unknown indices of refraction, it is not currently possible to present a convincing calculation of this ratio using the usual Mie scattering techniques. Even if a methodology is developed to theoretically derive the extinction-to-backscatter ratio, direct measurement would be necessary to confirm the approach.

This report deals with a description of a technique to directly measure the extinction-to-backscatter ratio of aerosols throughout the entire troposphere, and in addition, presents the results of two field measurements employing the described technique. Stratospheric results are also included since the physical and optical properties of the present high altitude aerosol layer are relatively well understood and might be considered a "calibration" or standard aerosol for comparison to tropospheric aerosols.

2. METHODOLOGY

The measurements described here utilize balloon borne instrumentation that simultaneously senses the local ambient aerosol backscatter as well as the total aerosol scattering. It should be noted that remote sensing type sensors are not utilized, and that vertical profiles are inferred from repeated sampling of local conditions as the instruments rise through the atmosphere. From our experience in the geographical region of the measurements it is known that the particulates have a relatively small absorbing component [Rosen et al., 1992]; consequently the total scattering is equivalent to the aerosol extinction for these aerosols. Thus, the working assumption is being made here that the total aerosol scattering and aerosol extinction are essentially equivalent within the accuracy of the measurements, and therefore the term "extinction-to-backscatter ratio" can be used interchangeably with the term "total scattering-to-backscatter ratio".

3. INSTRUMENTATION

The backscatter measurements were made with a previously developed backscattersonde. This device has been described by Rosen and Kjome, [1991] and is now in relatively widespread use. The absolute calibration of this device is obtained by comparing a ground based standard backscattersonde whose signal has been simultaneously intercompared with a flight version. Regions of negligible aerosol scattering (pure Rayleigh scattering) can often be identified in a subsequent backscatter sounding [Rosen and Kjome, 1991] thus allowing a post-flight absolute calibration of the ground standard. In practice, several ground standard backscattersondes are maintained to insure reliability and continuity, and scrutiny of the absolute calibrations are continually being made after each backscatter sounding.

The total scattering measurements were made with a unique balloon borne integrating nephelometer as described in a previous annual scientific report [Rosen, 1990]. Although the nephelometer is in widespread use, this is the first successful application to balloon soundings in which the relatively low free tropospheric aerosol concentration can easily be identified. The calibration of the instrument is achieved by comparing its response in ambient aerosol to an absolutely calibrated standard nephelometer sampling the same ambient aerosol.

In performing the calibration in this manner it must be recognized that the flight instrument has a small, but non-zero, value of constant background scatter from parts of the device itself (equivalent to wall scattering in the nephelometer calibration standard). This background value is determined at high altitude where the molecular and aerosol scattering are insignificant and is subtracted from the signal in the subsequent analysis. The consequence of errors in this determination are discussed below.

The standard nephelometer is calibrated by operating it in a large environmental chamber and pumping out the air as well as the aerosol. In this way the contribution to the signal from wall scattering can be accurately determined. When pure air (no aerosols) is allowed into the nephelometer at a known pressure and temperature the Rayleigh component can be inferred. Under normal operating conditions, the signal from the nephelometer contains a component from wall scattering, Rayleigh scattering, and aerosol scattering. Using the predetermined values of wall scattering and Rayleigh scattering, the aerosol component can be determined. It must be kept in mind that Rayleigh scattering is affected by the density of the air in the scattering chamber and that the wall scattering is not necessarily constant for extended periods of time. We have experimented with the stability of the ground based standard nephelometer by making several calibrations per week over an extended period, during which time the instrument was transported to and from the field site where calibrations with the flight instrument take place. It has been found that the wall component of scattering varies somewhat with time, but the Rayleigh component is constant within the statistical errors of measurement (about 5%). Best results are obtained by calibrating the standard nephelometer immediately before and after it is used to calibrate the flight nephelometer.

4. WAVELENGTH CONSIDERATIONS

Initially it was decided that it would be most useful to determine both the aerosol extinction and backscatter at the same wavelength so that the extinction-to-backscatter ratio would refer to a single wavelength. However, it is recognized that the ratio of extinction at one wavelength to the backscatter at another wavelength may also be useful. The mixed wavelength possibility will not be considered here.

5. WAVELENGTH INTERPOLATION

Since the present nephelometer utilizes only one wavelength (690 nm) the backscatter needs to be determined at this wavelength. A slight complication arises here in that the present backscattersonde only measures backscatter at 490 nm and 940 nm. However, we have found that the backscatter at 690 nm can probably be estimated to an accuracy of about 2% by interpolation using a power law relation between backscatter and wavelength and fitting the curve to the points at 490 nm and 940 nm. The accuracy of the interpolation method was evaluated theoretically by using a unique ensemble of tropospheric size distributions known to reproduce acceptably accurate tropospheric scattering properties [Rosen et al., 1992]. The backscatter calculated at 690 nm was first directly obtained by using Mie scattering calculations. Then the values of backscatter calculated at 490 and 940 nm were also calculated and used to find an interpolated value of backscatter at 690 nm. Both methods of obtaining the backscatter at 690 nm gave the same results within a few percent.

A direct experimental approach would be more certain in evaluating the accuracy of the above interpolation method. Undoubtedly, a large variety of tropospheric aerosols would need to be considered. Although we expect to do this in the future, it is an expensive and time consuming exercise, since both a ground based standard backscattersonde and a flight instrument need to be operated for an extended period before a reliable absolute calibration can be established.

6. RESULTS

Two balloon flights at Laramie were conducted on which both the backscattersonde and the nephelometer made simultaneous measurements. The results of these soundings are shown in Figures 1 and 2. It is convenient to present the backscattersonde results as a ratio of aerosol scatter to Rayleigh scatter, which will be referred to here as the aerosol backscatter ratio (ABKSR) and, as will be noted, is a dimensionless quantity. Similarly, the nephelometer results are presented as the ratio of (total angular) aerosol scatter to Rayleigh scatter for that instrument, which will be referred to here as the aerosol scatter ratio (ASR) and is also dimensionless. The ratio of the nephelometer to backscatter measurements corrected to 690 nm is shown in Figures 1 and 2 and denoted as ASR/ABKSR. Each data point (triangles for ascent, dots for descent) represents a 0.5 km average. Typical error bars are also shown and represent the uncertainty from statistical fluctuations as well as the calibration uncertainties. At altitudes above about 25 km the nephelometer response becomes quite noisy due to increasing statistical fluctuations in the small signal. Figure 1 shows the average value of ASR/ABKSR for data collected above 25 km, but as indicated, the uncertainty is quite large.

In order to obtain the ratio of aerosol scatter-to-backscatter, which under the above assumptions is equivalent to the aerosol extinction-to-backscatter ratio, the value of ASR/ABKSR must be multiplied by $8\pi/3$, which is the Rayleigh extinction-to-backscatter ratio. After this conversion the extinction-to-backscatter ratio will have units of steradians (sr). For example, Figures 1 and 2 suggest a value of ASR/ABKSR = 6 at the peak of the stratospheric aerosol layer. This infers a value of the extinction-to-backscatter ratio of about $6 * 8\pi/3 = 50$ sr at 690 nm for the present stratospheric aerosol.

7. DISCUSSION

The extinction-to-backscatter values implied by Figures 1 and 2 appear reasonably consistent with previous estimates used for the troposphere and stratosphere. Spinhirne et al., [1980] deduced values of 21 ± 6 sr for typical tropospheric values during their study period. Ferrare et al., [1992] have reported values of 50 to 70 sr for the stratospheric aerosol relevant to Figures 1 and 2. Similar values were inferred by Thomason and Osborn [1992] for stratospheric aerosols. The wavelength dependence of the extinction-to-backscatter ratio, which is useful for making consistency estimates with other available data, is discussed below.

A somewhat perplexing feature of the ASR/ABKSR profiles is the fact that they continue to increase in value for about 5 km above the stratospheric aerosol maximum. Conventional wisdom suggests that above the maximum the aerosol particle size should be decreasing with increasing altitude, since the larger particles settle out faster and are not as easily mixed to the higher altitudes. Theoretical scattering calculations indicate that this would lead to smaller, rather than larger, values of the ratio ASR/ABKSR above the aerosol maximum. However, the highest altitude point in Figure 1, which represents an average over about 6 km does, in fact, suggest a decreasing trend but is somewhat uncertain due to large statistical fluctuations associated with the weak signal at these altitudes.

The unexpected increase (or constant value) in the ratio of ASR/ABKSR above the stratospheric aerosol maximum might suggest an error in the flight nephelometer background and that its value is somewhat too low. Such an error of the magnitude required would tend to have a large influence on the high altitude points, but the low altitude data (below about 15 km) would remain relatively unchanged. The possibility of the nephelometer background being in error has been extensively examined, but the present value employed cannot be changed significantly within the constraints dictated by the profiles themselves. It is hoped that future research will be able to resolve this issue. For the present, however, the tropospheric values that are of prime consideration here are relatively insensitive to the uncertainty in background values.

It would be highly desirable to measure the extinction-to-backscatter ratio at multiple wavelengths. During the next year it is expected that the nephelometer will be modified so that the extinction-to-backscatter for at least two wavelengths can be determined. Until modifications of the instrument can be made, it is useful to consider a theoretical approach to estimating the wavelength dependence of the extinction-to-backscatter ratio. The results of such calculations are shown in Figure 3 for both the troposphere, and for comparison, the stratosphere. The tropospheric curve represents an average of results obtained from an ensemble of size distributions measured throughout the boundary layer and free troposphere [Rosen et al., 1992]. As previously mentioned the size distributions belonging to this ensemble are unique in that they are known to be consistent with the correct optical properties as simultaneously and independently measured. The curve for stratospheric aerosols shown in Figure 3 refers to highly volcanic conditions (for about 1 year after a major eruption) and is based on size distributions given by Rosen and Hofmann [1986] and Deshler et al. [1992].

8. CONCLUSIONS

The above described method for directly measuring the extinction-to-backscatter ratio appears to be an operationally realistic procedure producing realistic results that are consistent with previous estimates. Theoretical calculations of the color dependence of the extinction-to-backscatter ratio for tropospheric aerosols shows a relative insensitivity to wavelength. Experimental verification of this result will take place after the nephelometer has been modified. Since almost no reliable measurements of extinction-to-backscatter are available for atmospheric aerosols, it is now necessary to conduct a reasonably extensive effort to make surface based measurements of this parameter under a variety of conditions. Such a data base is needed for comparison and evaluation of results obtained throughout the boundary layer and free troposphere.

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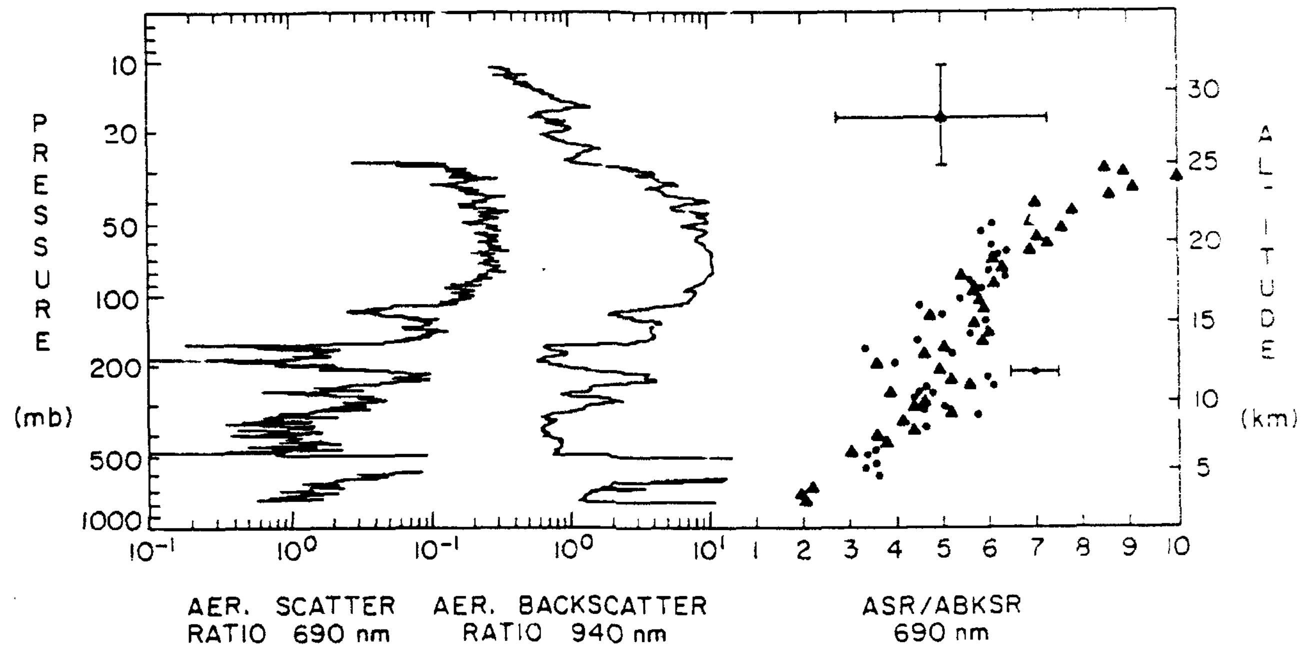

Figure 1. Simultaneous backscatter and nephelometer measurements made at Laramie on 13 July 1992 during a single balloon sounding. The profile on the right side (ASR/ABKSR) is the ratio of the two profiles on the left side and under certain assumptions is equivalent to the aerosol extinction-to-backscatter ratio. See text for details.

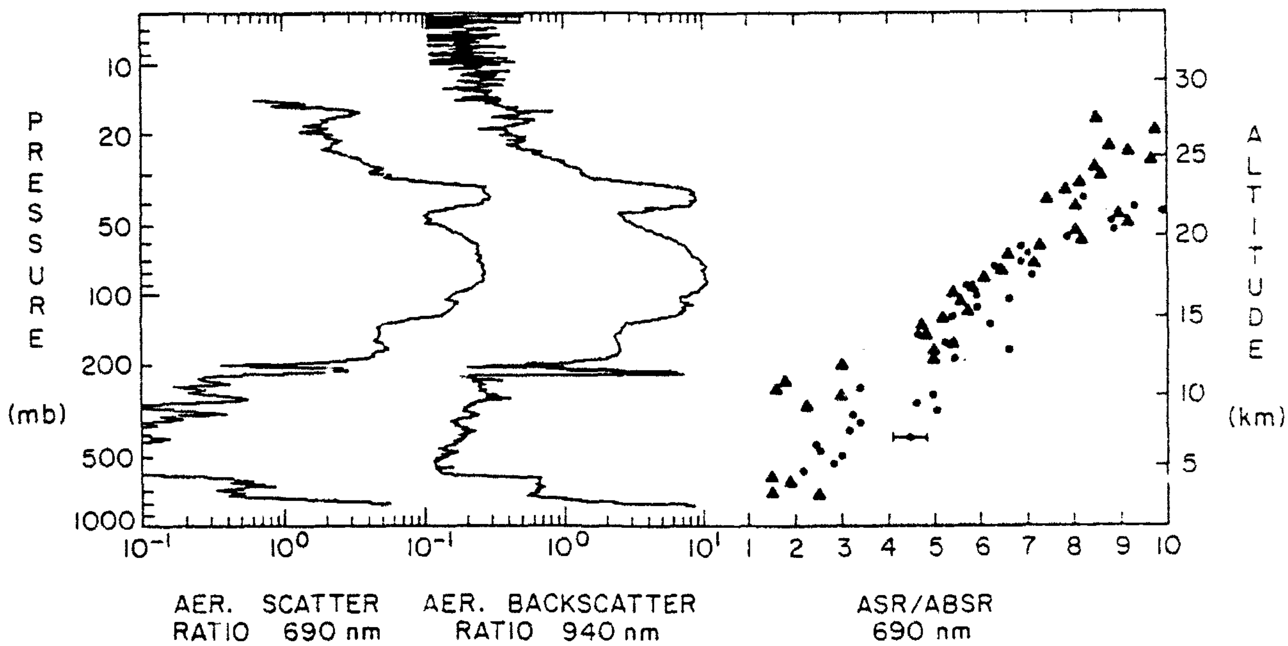

Figure 2. Simultaneous backscatter and nephelometer me..surements made at Laramie on 28 September 1992 during a single balloon sounding. The profile on the right side (ASR/ABKSR) is the ratio of the two profiles on the left side. See text for explanation of the aerosol extinction-to-backscatter ratio implied by the ASR/ABKSR profile.

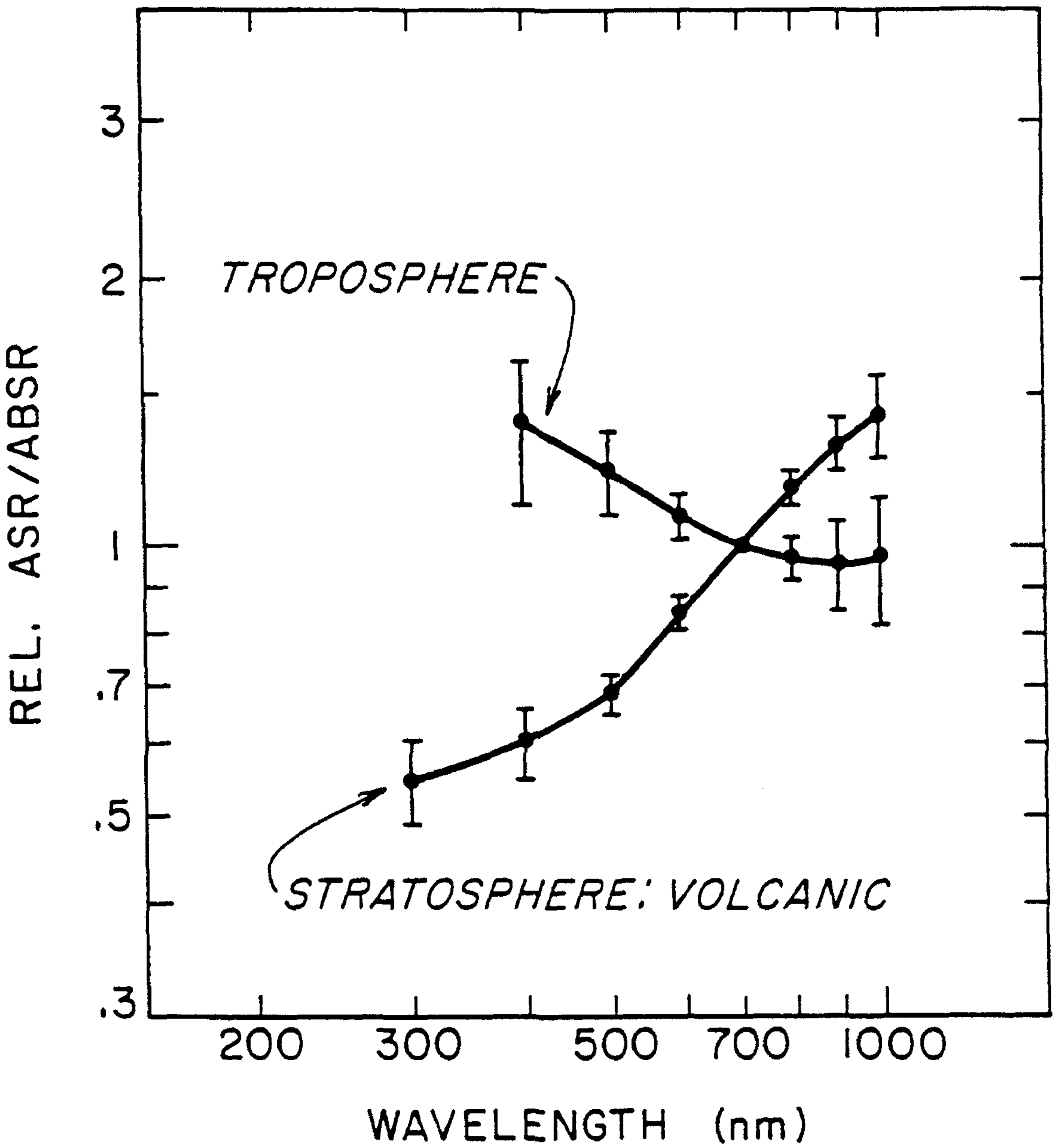

Figure 3. The wavelength dependence of the aerosol extinction-to-backscatter ratio for tropospheric and stratospheric aerosols as based on theoretical calculations using measured size distributions.