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**AEROSOL OPTICAL PROPERTIES
OF THE FREE TROPOSPHERE :
TROPOSPHERIC BACKSCATTER CLIMATOLOGY**

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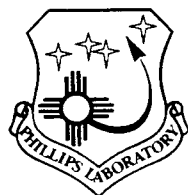


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13. ABSTRACT (Maximum 200 words)

A unique ensemble of aerosol sensors (backscatter sondes, nephelometers and particle counters) has been assembled during the course of this research to obtain new measurements relating to the optical properties of aerosols in the atmosphere, especially in the free troposphere. A knowledge of the aerosol extinction-to-backscatter ratio has been greatly enhanced as a result of this project and the inference of representative values along with the range of variation is now possible. Agreement between the optical model results and actual measurements appears to be quite satisfactory. An initial climatology of aerosol backscatter in the free troposphere has been developed and is in general agreement with results and inferences from global remote sensing instruments. However, the data from remote sensors may indicate a larger influence of volcanic aerosols on the upper troposphere than actually exists. Further work with high resolution soundings is needed to fully resolve this issue.

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SUMMARY

The principal results and progress in the areas of study addressed by this research can be summarized as follows:

Development and construction of a balloon borne open cavity nephelometer.

Construction of a closed cavity nephelometer and associated vacuum chamber for making absolute calibrations.

Development and construction of a closed cavity backscatter nephelometer for making absolute calibrations of the backscattersonde.

Configured backscattersonde, open cavity nephelometer and particle counter into a complete system for balloon flight application.

Conducted a total of more than a dozen balloon soundings involving the open cavity nephelometer, the backscattersonde as well as particle counters.

Participated in an unusual target-of-opportunity program (Front Range Lidar, Aircraft and Balloon experiments - FRLAB) to study aerosol optical properties of the free troposphere.

Performed comprehensive analysis of the FRLAB results.

Made extensive measurements of the extinction-to-backscatter ratio in the boundary layer to determine typical values and variability for comparison with values obtained in the free troposphere.

Performed optical model calculations using measured size distributions as a basis and compared results with concurrent measurements of backscatter and total scatter. As a byproduct, a representative ensemble of "working" size distributions was identified relevant to the free troposphere and boundary layer.

Developed a "climatology" describing the free tropospheric backscatter over Laramie, which is thought to be typical of mid-latitude, continental conditions.

Performed a critical review and assessment of the SAGE I/II tropospheric results in relation to our findings. Studies directed at the validation and interpretation of SAGE I/II data are considered essential because satellite based observations are the only realistic means of global exploration of the free troposphere.

1. INTRODUCTION

The primary goal of the work conducted under this effort was to obtain and analyze concurrent measurements of diverse optical properties relating to aerosols in the free troposphere. Specifically, the parameters measured were total scattering (into all angles), backscatter, and size distribution. These three parameters furnish a very effective and sensitive basis for evaluating the accuracy of optical model calculations based on size distribution and index of refraction. Most of the measurements reported here were made from regular balloon soundings at Laramie and provide, as an important byproduct, a preliminary climatology of the free troposphere aerosol optical properties. Although the free troposphere was the focus of this study, the planetary boundary layer and stratosphere could not be completely neglected, because aerosols from these regions can potentially affect large portions of the atmosphere.

This report deals primarily with the presentation of results obtained since the last annual progress report. Three main areas of advancement will be described: completion of the extinction-to-backscatter measurements supported by corresponding optical model calculations, a climatology of the free troposphere backscatter over Laramie (which is probably typical of a continental mid-latitude site), and a comparison of our results with those from global satellite observations (SAGE) of the free troposphere. This last area of study presents a critically-needed, independent verification of new tropospheric results coming from the satellite measurements.

2. INSTRUMENTATION

2.1. The Backscattersonde

The backscattersonde is a simple and relatively new balloon borne sensor. It employs a quasi-collimated beam from a xenon flash lamp and senses the light locally backscattered at two wavelengths (940 and 490 nm.) The vertical resolution is about 30 meters as determined by the balloon rise rate and frequency of flashes from the lamp. Standard meteorological parameters such as pressure, air temperature and relative humidity, as well as ozone, are also determined by the instrument. A more complete description of the instrument and its calibration is given by Rosen and Kjome [1991]. A comparison of the backscattersonde with lidar is given by McKenzie et al., [1994] and by Rosen et al., [1992].

2.2. The Nephelometer

The balloon borne nephelometer has been described in a previous report [Rosen 1990] and employs essentially the same scattering geometry as commercially available instruments [Alquist and Charlson, 1969]. The flight instrument was calibrated against a specially constructed standard laboratory nephelometer capable of operating in an environmental chamber under partial vacuum conditions. In this way, the standard nephelometer could be calibrated in an absolute sense without the need for calibration gases whose properties are somewhat uncertain [Bodhaine, 1979].

2.3. Particle Counter

The optical particle counters employed in this study are similar to those first described by Rosen [1964] with later changes described by Deshler et al. [1992].

3. THE EXTINCTION-TO-BACKSCATTER RATIO

The extinction-to-backscatter ratio is a crucial but poorly known parameter needed in the processing of many types of remote sensing data. In addition, this measured ratio provides an excellent focus for a stringent evaluation of the internal consistency of optical models based on size distribution calculations. In a previous report a methodology for making a relatively direct determination of the extinction-to-backscatter ratio was described [Rosen, 1992] and made use of the fact that aerosol extinction and backscatter are very nearly identical for low absorption particles. Vertical profiles of the extinction-to-backscatter ratio obtained under this program have been presented earlier [Rosen, 1992] but without the companion optical model calculations. This section deals with a description and presentation of the companion model calculations predicting the extinction-to-backscatter ratio, as well as a description of new field measurements which establish a typical value for the extinction-to-backscatter ratio (or range of typical values) in the planetary boundary layer.

3.1. Ground Based Measurements

Before presentation of the optical model calculation it would be convenient to first consider the results of a relatively extensive effort to measure the extinction-to-backscatter ratio at a field site near Laramie. This direction of research was considered important and necessary primarily because it supported the assessment and credibility of the balloon borne measurements. In a secondary sense, it was recognized that we had at our disposal the means of making the most extensive and reliable extinction-to-backscatter measurements ever reported.

The instrumentation employed for the boundary layer study of extinction-to-backscatter ratio was the backscatter sonde and the closed cavity nephelometer operating in close proximity. A continual effort was made to keep both instruments in good calibration. Since the two instruments do not operate at exactly the same wavelengths it was necessary to interpolate the backscatter measurements (at 490 and 940 nm) to one of the wavelengths utilized by the nephelometer (690 nm). As previously discussed [Rosen, 1992], only a small uncertainty of about 2% is expected in the interpolated value and is not significant compared to the overall measurement uncertainty of about 10%.

On more than 30 evenings between 13 May 1993 and 14 March 1994 measurements were obtained at the field site near Laramie and are representative of a variety of wind conditions and directions. Continuous measurements were not possible because the instrumentation was not designed to operate unattended. In the study reported here, no attempt has been made to correlate the nature of the results with meteorological parameters.

The results of the measurements made near Laramie are shown in Figure 1, which for comparison, also illustrates results obtained on 3 target-of-opportunity occasions (FRLAB I/II [Rosen et al., 1992] and ALIVE II [Hoidale et al., 1991]). For presentation in the figure, the measurement results themselves have been normalized by the equivalent clean air scattering value so that an immediate appreciation for the magnitude of the aerosol scattering relative to pure molecular scattering can be obtained. The lines labeled S/B give the ratio of the aerosol scatter ratio (over 4π solid angle) to the aerosol backscatter ratio. As discussed above, the value for S/B is the same as the extinction-ratio-to-backscatter-ratio ratio for low absorbing aerosols and in this format is a dimensionless number. In contrast, the extinction-to-backscatter ratio used by some researchers has dimensions of sr^{-1} and is obtained from

the dimensionless numbers through multiplying by $8\pi/3$, which is the molecular extinction-to-backscatter ratio.

The mean S/B value and standard deviation for the points shown in Figure 1 is $2.54 \pm .53$, or 21.3 ± 4.5 sr. This result is similar to the value of 21 ± 6 sr obtained by Spinhirne et al., [1980] for a boundary layer aerosol.

It was previously shown on theoretical grounds through optical modeling of tropospheric aerosols [Rosen, 1992], that the extinction-to-backscatter ratio is not expected to be strongly dependent on wavelength over the range of interest. Experimentally it was possible to verify this prediction in the wavelength region between 690 and 525 nm by using the data from the 525 nm nephelometer channel and performing an analysis similar to that illustrated in Figure 1. The average value of the extinction-to-backscatter ratio at 525 nm was found to be $2.77 \pm .75$ or 23.2 ± 6.0 sr. The factor increase between 690 and 525 nm is only 9% and consistent with that predicted by the model.

3.2. Balloon Borne Measurements

Vertical profile measurements of the extinction-to-backscatter ratio at 690 nm have been presented in an earlier report [Rosen, 1992]. However, in this preliminary work the optical modeling calculations had not been completed, and therefore it was not possible to compare the measurements with values predicted utilizing the concurrently measured aerosol size distributions. Figures 2 and 3 contain the results of the optical model calculations and their comparison to the actual measurements. Also added to the figures is the range of values of extinction-backscatter ratio observed at the surface.

3.3. Optical Model Calculations

The size distributions utilized in the optical model calculations relating to Figures 2 and 3 are based on 1 km averages taken on five balloon soundings covering the period from June to October 1992. The error bars shown in the figures represent one standard deviation in the mean of the five values used. Since the stratospheric size distribution was probably fairly stationary during this period, the error bars probably can be interpreted as a reflection of the inherent reproducibility of the size distributions and the consequent scattering calculations.

Considering the magnitude of uncertainties, the agreement between the measured and calculated values shown in Figures 2 and 3 is quite satisfactory. The higher values of extinction-to-backscatter ratio seen in the upper troposphere in Figure 2 probably are due to transport of stratospheric air to lower altitudes as suggested by the accompanying scatter and backscatter ratio profiles. The ozone profile obtained in this sounding (not illustrated) shows a strong positive correlation with the layered structure in the upper troposphere, again suggesting that this region was strongly influenced by stratospheric air at the time of the sounding. Such transport layers are not present in Figure 3, and the corresponding extinction-to-backscatter ratio is lower, which is probably more typical of tropospheric conditions.

The concurrent measurements that have gone into the making of Figures 2 and 3 are truly unique and only made possible by support of this research contract. It is not likely that the casual reader will appreciate the amount of effort and persistence needed to produce these two figures.

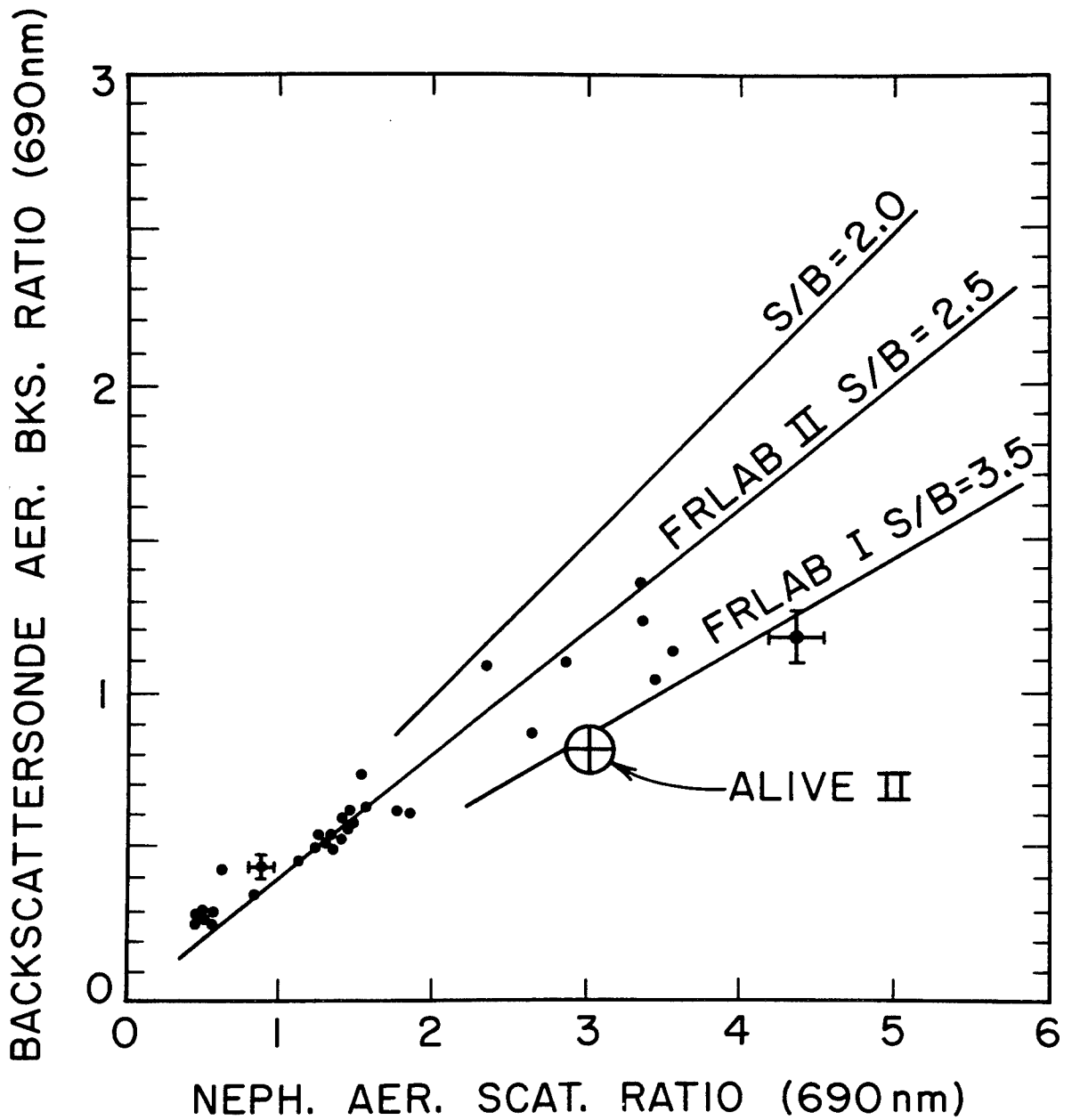


Figure 1. The results of simultaneous backscatter and nephelometer measurements at a field site near Laramie (dots). Typical error bars are illustrated for two data points. The straight lines labeled with values of S/B refer to constant values of aerosol scatter ratio divided by aerosol backscatter ratio. The lines labeled FRLAB I and FRLAB II and the point labeled ALIVE II refer to results obtained on target-of-opportunity occasions for single air masses (see text for details).

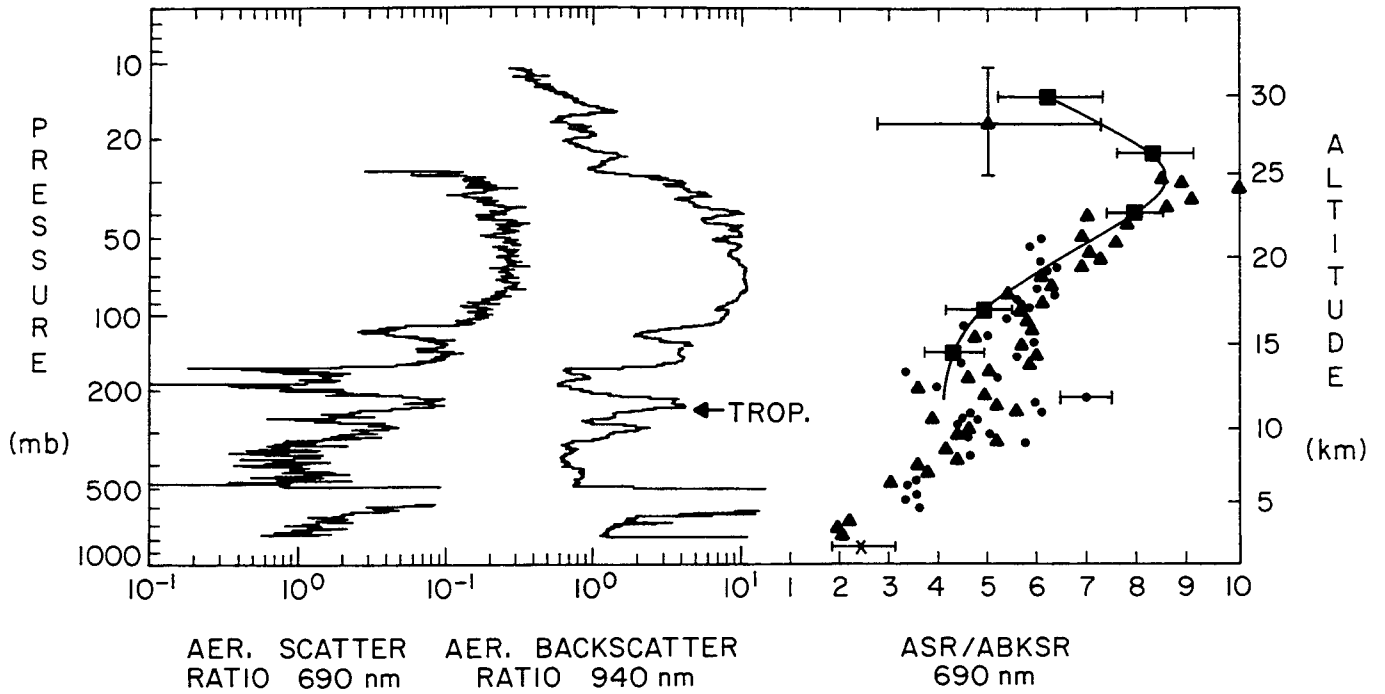


Figure 2. Simultaneous backscatter and nephelometer measurements made at Laramie on 13 July 1992 during a single balloon sounding. The profile of triangles (ascent) and dots (descent) on the right side (ASR/ABKSR) is the ratio of the two profiles on the left side, and under certain assumptions is equivalent to the aerosol extinction-to-backscatter ratio (see text for details). The solid line on the right represents predicted values based on calculations utilizing concurrently measured size distributions. Typical error bars are illustrated.

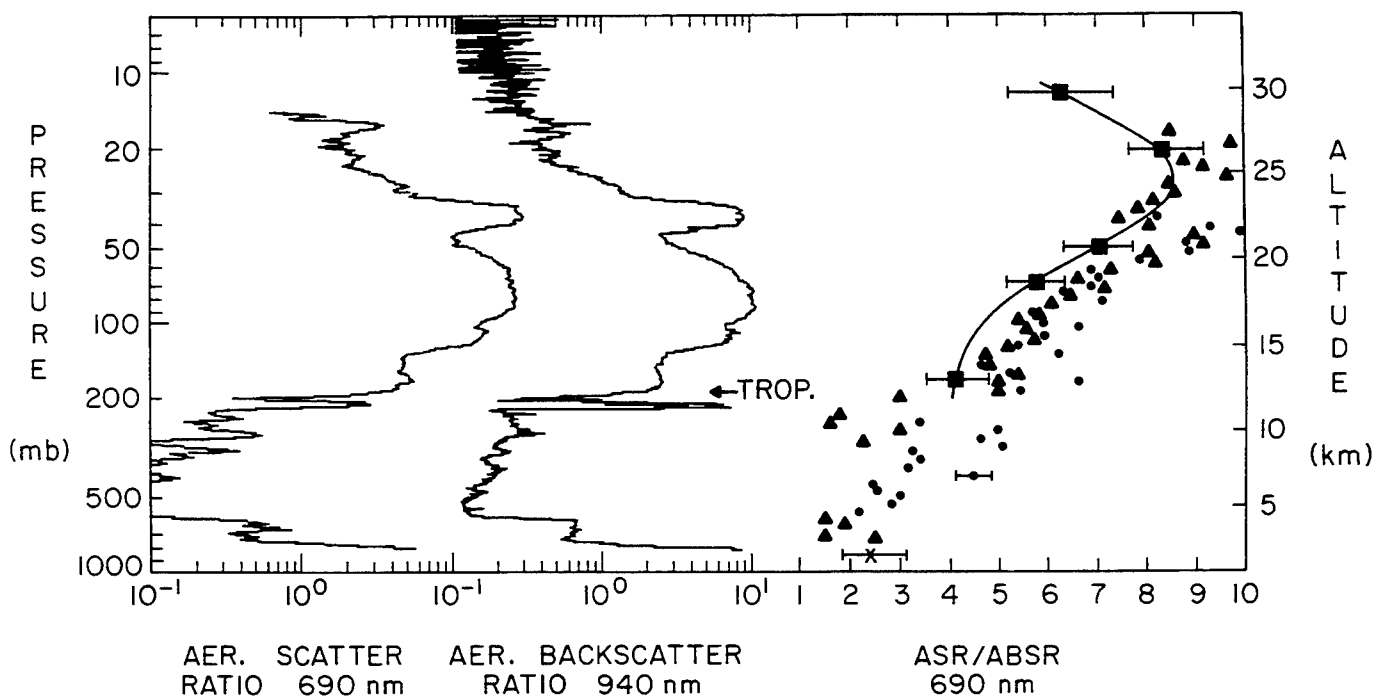


Figure 3. Same as Figure 2 except for 28 September 1992.

4. TROPOSPHERIC BACKSCATTER CLIMATOLOGY

An important aspect of this program was to obtain an initial data base describing the mean aerosol optical properties of the free troposphere and their expected range of variation. As argued above, it is also important to compare the aerosol conditions in the free troposphere with those of the planetary boundary layer and the stratosphere. This report focuses on backscatter as an indicator of aerosol optical properties because a knowledge of the absolute value and variations is essential to the calibration of remote sensing techniques such as lidar. The measurements reported here were made with the backscatter sonde, and unless otherwise stated, refer to a wavelength of 940 nm.

4.1. Annual Average

An overall summary of the backscatter measurements is shown in Figure 4 and represents non-cloud data taken from August 1991 to May 1994. The large range of stratospheric values is due to the injection and decay of debris from the Mt. Pinatubo eruption in June 1991. Individual data points for pre-Pinatubo conditions are not shown directly, but the envelope of maximum values is indicated by the solid line. An initial assessment of this figure might suggest that the concentration of free tropospheric aerosols could be highly influenced by the magnitude of aerosols in the stratosphere (depending on volcanic conditions) and planetary boundary layer. Nevertheless, there appears to be a predominant peaking in the free tropospheric aerosol backscatter ratio at about 0.2, with values less than 0.1 rarely occurring.

4.2. Seasonal Variations

The backscatter measurements illustrated in Figures 5 and 6 represent the first step in an effort to identify seasonal variations. Even a casual comparison of the two figures indicates a significant enhancement of free tropospheric aerosols during the defined "Spring season". The possible reason for the enhancement will be discussed below. The typical free tropospheric values of aerosol backscatter ratio during the Fall season is about 0.2 and is apparently responsible for the peaking in values discussed above.

4.3. Identifiable Regions of Characteristic Size

An elementary indication of the particle sizes can be obtained by utilizing the two color data obtained in the backscatter measurements. To this purpose a color ratio has been defined as the aerosol backscatter ratio at 940 nm divided by the aerosol color ratio at 490 nm. Through experience and theoretical calculations three main size ranges can be identified: particles with diameters typically less than 0.05 μm give color ratios less than about 4.0; particles in the size range from about 0.10 to 1.0 μm diameter produce color ratios from about 4.0 to 8.0; particles larger than about 1 μm diameter give color ratios greater than about 10 with a maximum possible around 15. For example, at the edge of clouds, where the instrument readings are not saturated, the mean value of the color ratio is 13.7 with a standard deviation of 2.0 (cloud particles are usually thought to be significantly larger than 1 μm). Blowing dust and smoke conditions produce color ratios of about 10. Optical model calculations of stratospheric aerosols based on measured size distributions give color ratios of about 6 ± 1 .

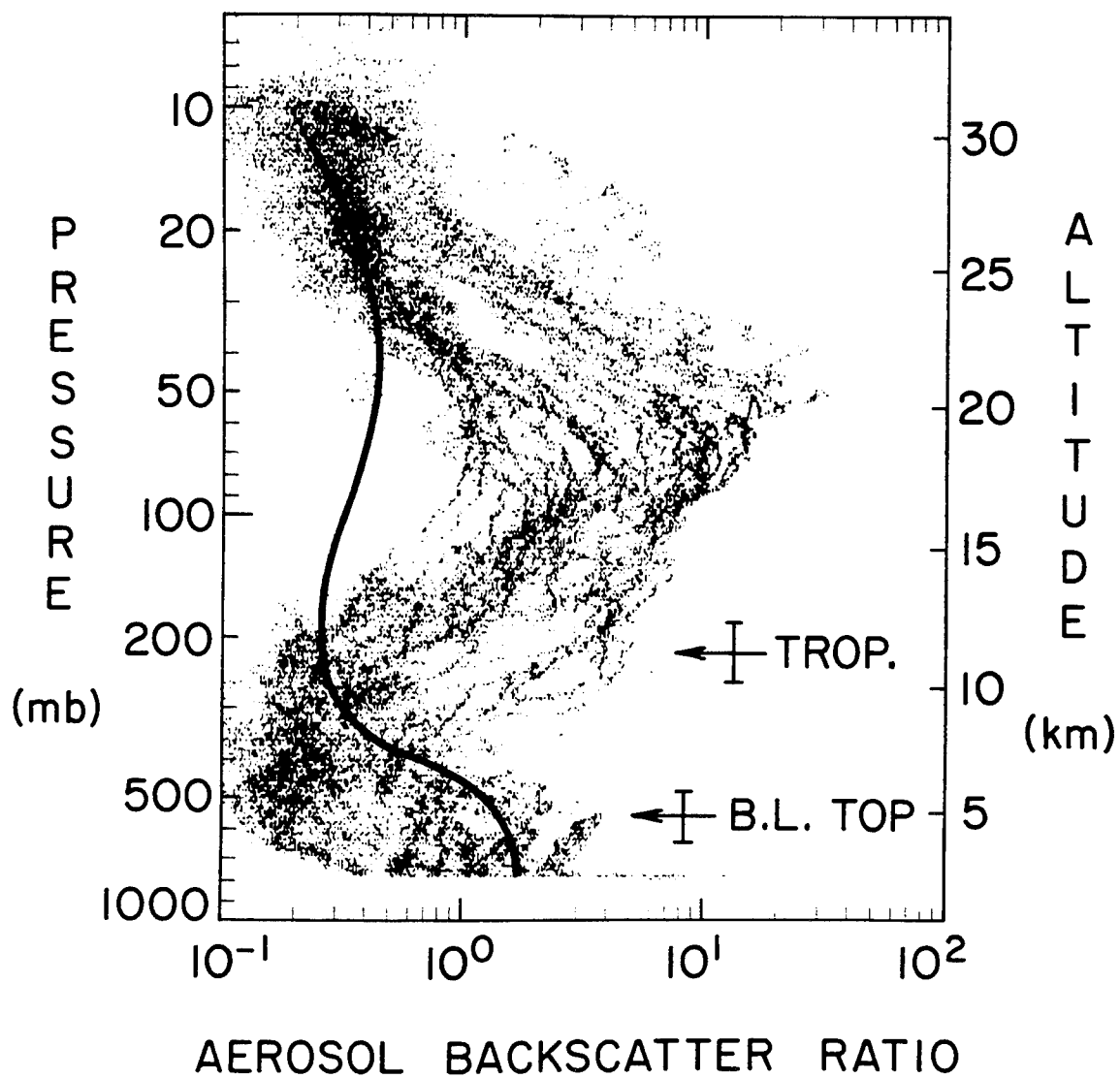


Figure 4. Results of backscatter measurements over Laramie for the period of 29 August 1991 to 16 May 1994. The average tropopause height and planetary boundary layer top (BL) are noted by arrows. The solid line represents the maximum of the envelope of pre-Pinatubo profiles.

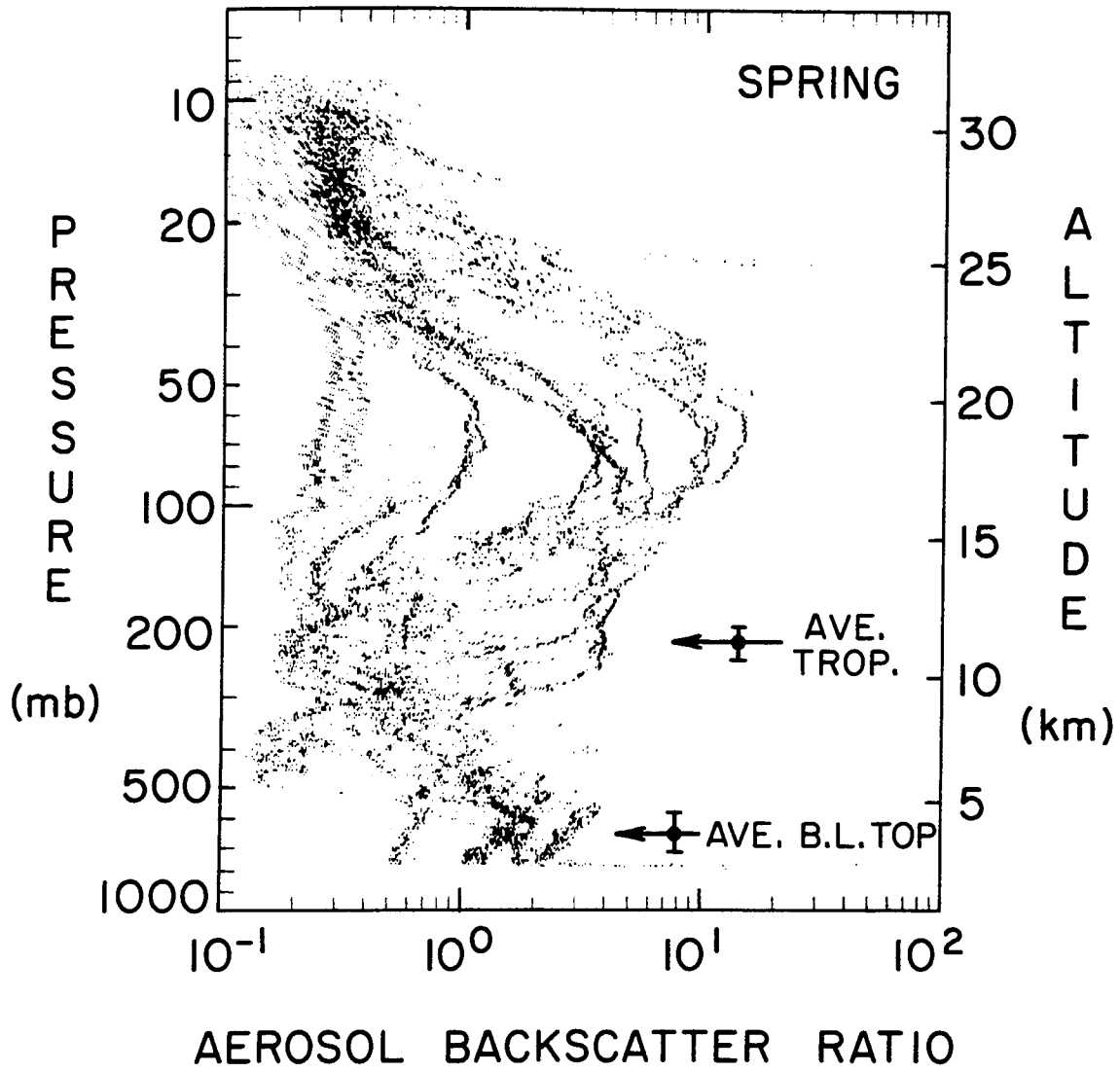


Figure 5. Same as Figure 4 except refers to data obtained from March to July.

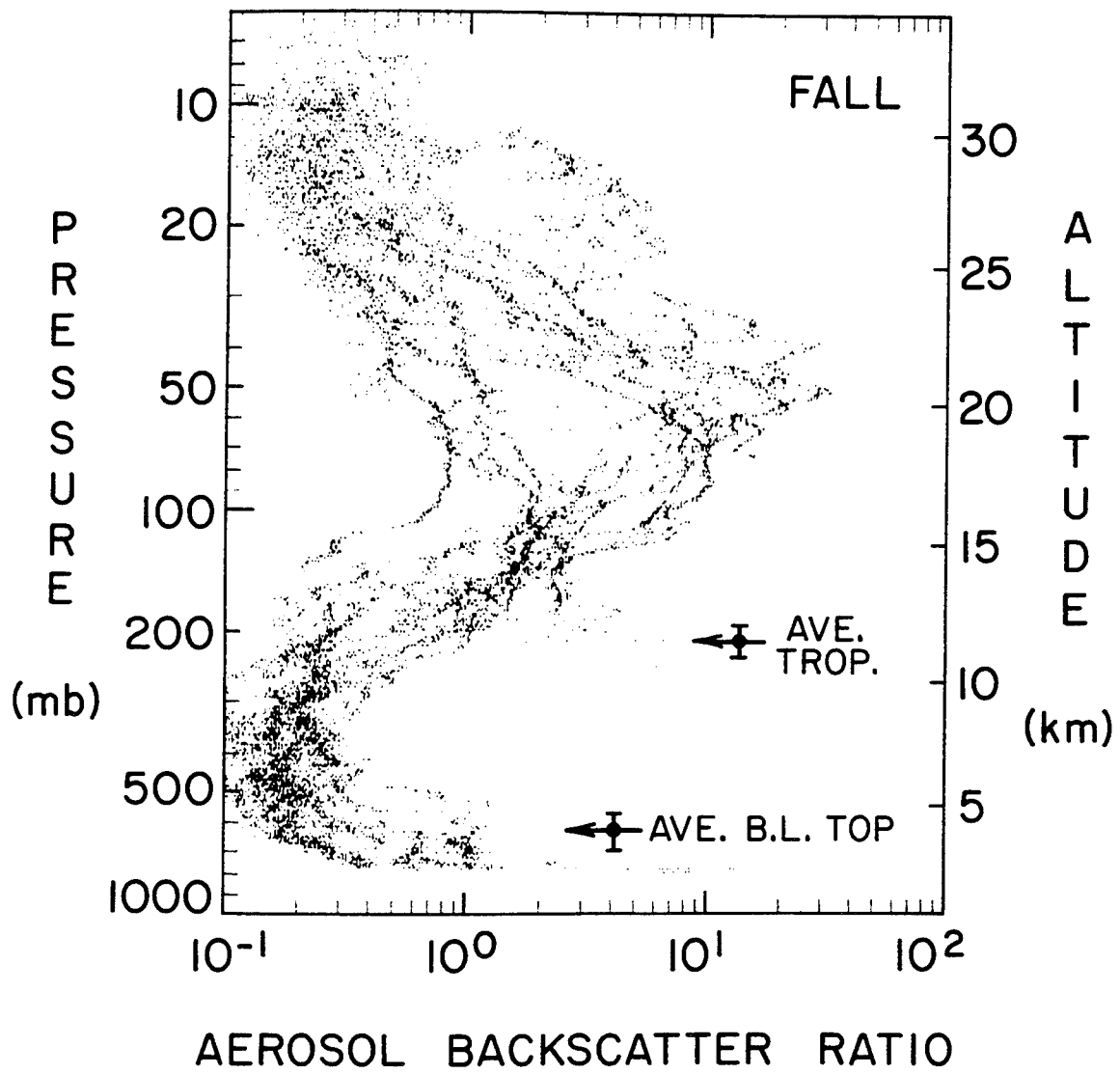


Figure 6. Same as Figure 4 except refers to data obtained from September to January.

Color ratios lower than about 3 are rarely observed in the natural atmosphere because these particles are too small to produce measurable aerosol scattering regardless of the quantity present.

Figure 7 illustrates the color ratios observed for the data plotted in Figure 4. Also contained in the figure at the bottom is the mean and one standard deviation of the color ratio values obtained during the efforts to measure extinction-to-backscatter ratio as described above. The solid lines represent a possible interpretation of source regions with identifiable color ratios. Even though cloud observations have been removed, some large color ratios are still present in the free troposphere, which are probably associated with near-cloud conditions or perhaps subvisible cloud layers.

The general color ratio values in Figure 7 indicate that free tropospheric aerosols tend to be smaller than the boundary layer aerosols and the post volcanic stratospheric aerosols. However, there are large variations in the color ratio, undoubtedly influenced by the aerosols from above and below. It is interesting to note the similarity in the mean color ratio of boundary layer aerosols and stratospheric aerosols, which indicates a roughly similar effective size.

4.4. Atmospheric Optical Depth Considerations

In making optical depth measurements from ground level, it would be very useful to know the contribution of aerosols in the planetary boundary layer. The present data set indicates that approximately 75% of the total backscatter from the troposphere comes from the boundary layer for springtime conditions. The contribution drops to 50% for Fall season. These percentages are probably very similar to that expected for extinction. Thus, optical depth measurements from ground level stations may be highly controlled by aerosols in the boundary layer and not be representative of most of the atmospheric volume above the observation site. High altitude sites would potentially be less affected, but even at Laramie (2.16 km altitude) the boundary layer contribution is overwhelming.

5. COMPARISON OF FINDINGS WITH SAGE RESULTS

Much of what is known and will be discovered concerning free tropospheric aerosols can be attributed to satellite based remote sensors, especially the SAGE I/II and SAM II instruments [Kent et al., 1995; Kent et al., 1994; Kent et al., 1991; Kent et al., 1988]. From a scientific point of view, it is very important that the present body of knowledge accumulated by what amounts to a single source be independently scrutinized and verified wherever possible. The following discussion is presented in response to such considerations.

5.1. Constraints on the Comparison

In making comparisons with the satellite data it is important to realize that the remote observations of the troposphere are possible only under extensive cloud free regions and that the effective operational minimum altitude is about 6.5 km, which is usually well above the planetary boundary layer. Therefore, the satellite data may be biased toward conditions that exist only during horizontally extended clear sky conditions in the upper troposphere. The backscattersonde measurements are made during a larger variety of atmospheric conditions but not during the occurrence of precipitation or heavy cloud cover. Thus, both the satellite

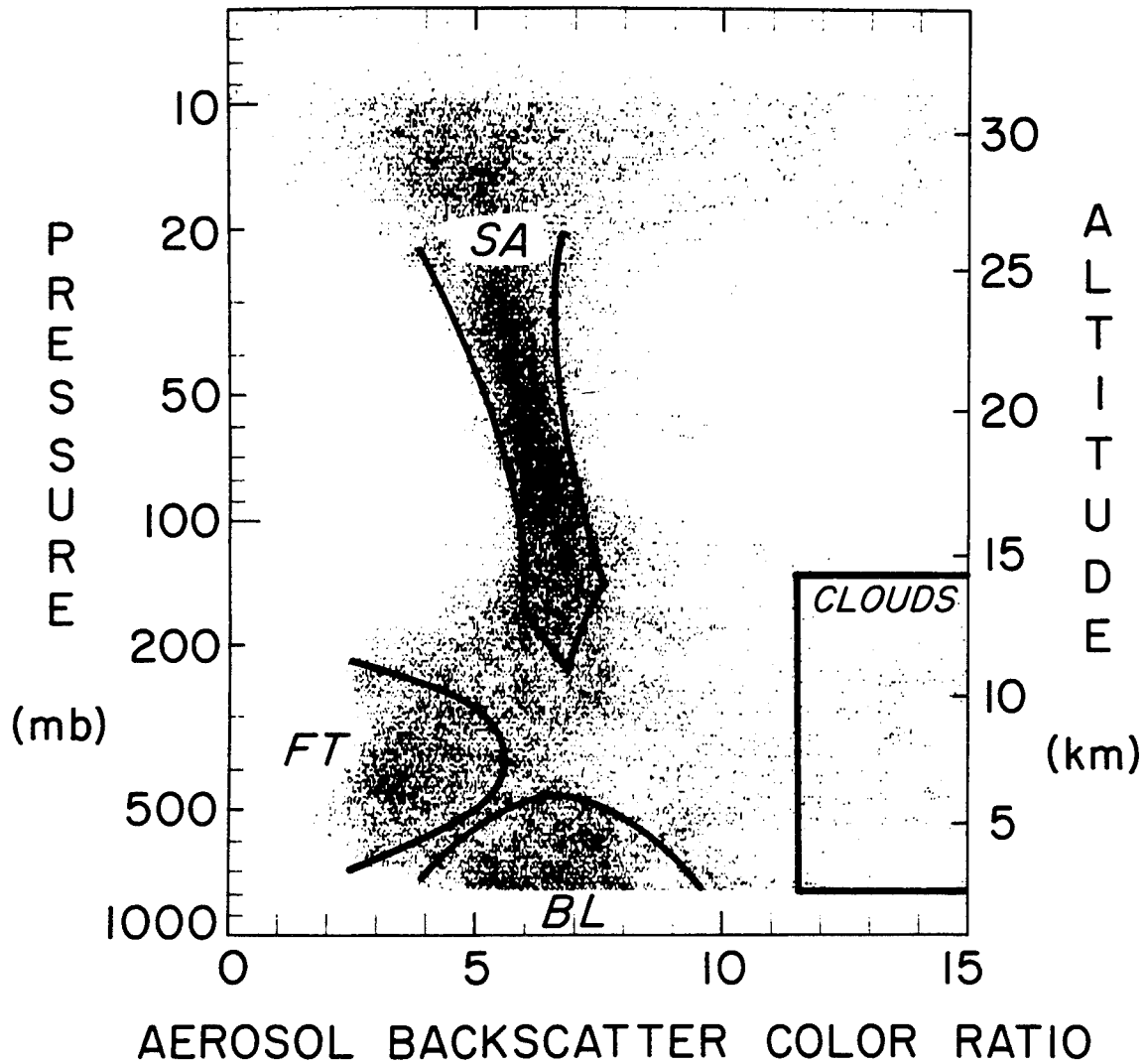


Figure 7. The color ratio values corresponding to the data shown in Figure 4. Solid lines surround suggested characteristic regions. Key: SA=stratospheric aerosol, FT=free troposphere, BL=planetary boundary layer.

and backscattersonde observations do not represent random samples of the atmosphere and may not be strictly comparable data sets. Nevertheless, the working assumption being made here is that both sets of observations should capture the essence of atmospheric aerosol behavior.

5.2. Mass Loading in the Free Troposphere

Perhaps the simplest comparison between the two types of observations is the inferred mass loading of the free troposphere. The published typical values derived from the satellite measurements are about $0.35 \pm$ (value not given) ug/m^3 at mid northern latitudes in the 6 to 10 km altitude range. The free tropospheric mass loading from the backscattersonde measurements can be estimated using a conversion factor such as that illustrated in Figure 8, which is based on an ensemble of "verified" size distributions [Rosen et al., 1992]. Using the typical value of free tropospheric backscatter as discussed above, Figure 8, and converting the mixing ratio (ppbm) to concentration the backscattersonde derived value is $0.35 \pm .1 \text{ ug}/\text{m}^3$, where the uncertainty represents one standard deviation of observed values. Considering the probable error in making such calculations and conversions, the apparent good agreement between the two methods is undoubtedly fortuitous, and agreement within a factor of two should perhaps be considered exact.

5.3. Seasonal Variations

Seasonal variations of free tropospheric aerosols at mid latitudes observed by the satellite sensors also indicate a significant Spring maximum and a Fall minimum in agreement with the backscattersonde data set. The exact months for which the extreme values occur may still be open to question. For both sets of observations, the magnitude of the seasonal variation (about a factor of two) is also in agreement.

Even though the SAGE data does not extend to altitudes less than 6.5 km, it was deduced that there is movement of material into the upper troposphere from below, particularly under springtime conditions. The backscattersonde data illustrated in Figures 4, 5, and 6 also suggest that a significant amount of aerosol moves into the free troposphere, especially during the Spring season.

5.4. Volcanic Influence

It has also been concluded from an analysis of the satellite data that volcanic stratospheric aerosols move into the upper troposphere and measurably disturb the normal conditions. A casual inspection of Figure 4 certainly would seem to support this observation. However, on closer inspection when the tropopause height for each sounding is employed (rather than an overall average) the stratospheric aerosol in most cases seems to have little affect on the aerosol concentration just below the tropopause. As an illustration, Figure 3 (which is typical of most soundings) shows a drop in aerosol concentration to background values just below the tropopause. However, a small percentage of soundings show layered aerosol structure in the upper troposphere, such as illustrated in Figure 2. These layers almost always contain ozone mixing ratios typical of the stratosphere and therefore are of probable stratospheric origin. Thus, in this small percentage of cases, the stratospheric aerosol has an easily observable affect on the upper troposphere. The satellite analysis probably overestimates the influence of volcanic stratospheric aerosols in the upper troposphere by using average troposphere heights rather than the specific heights.

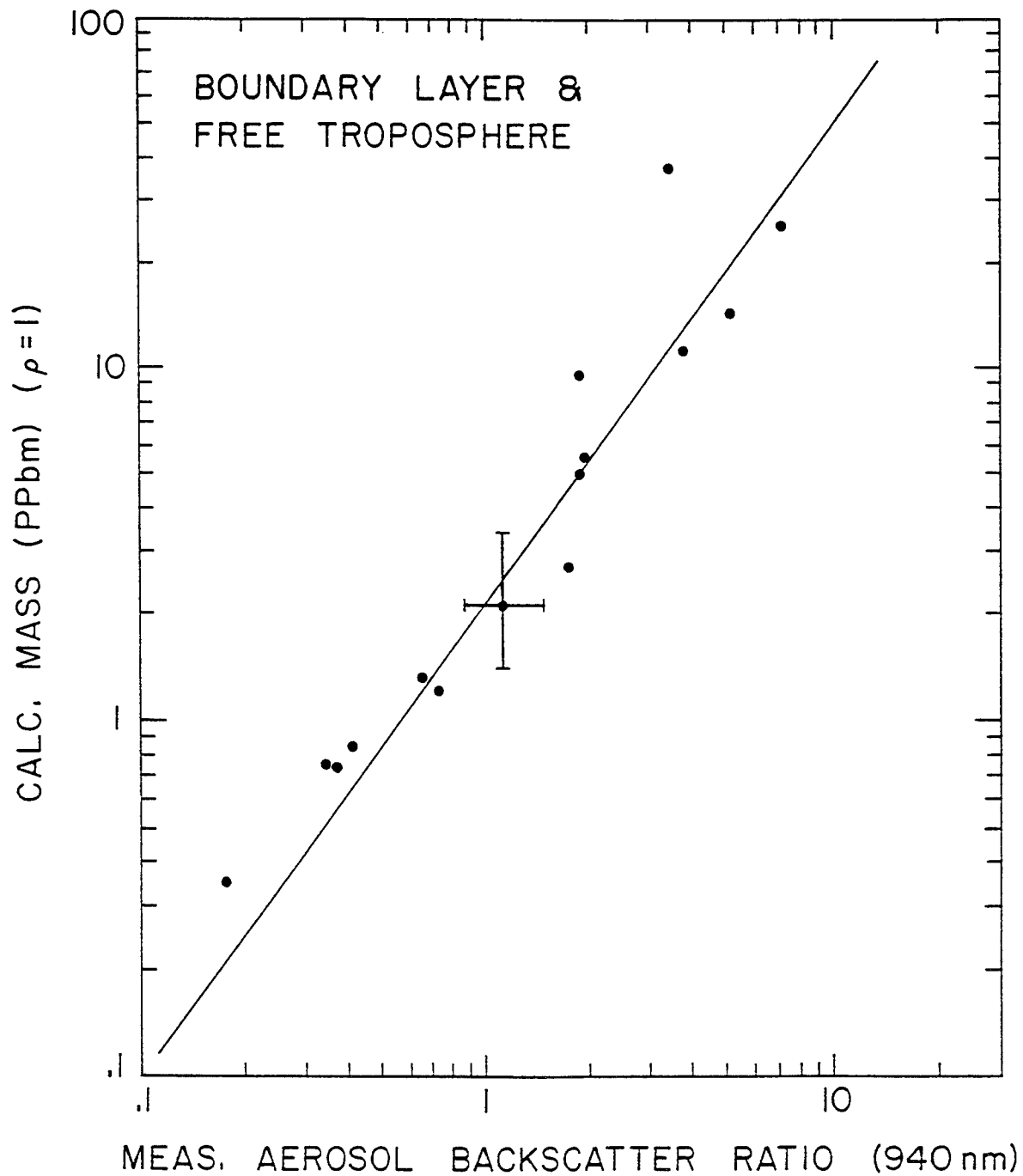


Figure 8. Relationship between backscatter and mass loading for tropospheric aerosols encountered during FRLAB I and II [Rosen et al., 1992]. A typical error bar is illustrated for one point only.

6. CONCLUSIONS

A unique ensemble of aerosol sensors has been assembled during the course of this research to obtain new measurements relating to the optical properties of aerosols in the atmosphere, especially in the free troposphere. A knowledge of the aerosol extinction-to-backscatter ratio has been greatly enhanced as a result of this project, and the inference of representative values along with the range of variation is now possible. Agreement between the optical model results and actual measurements appears to be quite satisfactory. An initial climatology of aerosols in the free troposphere has been developed and is in general agreement with results and inferences from global remote sensing instruments. However, the data from remote sensors may indicate a larger influence of volcanic aerosols on the upper troposphere than actually exists. Further work with high resolution soundings is needed to fully resolve this issue.

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