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Project Director: Dr. E. M. Patterson  School/Dept. Geophysical Sciences
Sponsor: Electronic Systems Division; Air Force Systems Command; Hanscom AFB, MA.

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Award Period: From 1/25/82 To 12/31/84 (Performance) 3/31/85 (Reports)
Sponsor Amount: $153,017 ($40,000 obligated through 9/30/82) Contracted through: GTRI/GIT

Title: Optical Absorption Characteristics of Aerosols

ADMINISTRATIVE DATA
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Defense Priority Rating: None  Security Classification: None

RESTRICTIONS
See Attached SFRL Supplemental Information Sheet for Additional Requirements.
Travel: Foreign travel must have prior approval – Contact OCA in each case. Domestic travel requires sponsor approval where total will exceed greater of $500 or 125% of approved proposal budget category.
Equipment: Title vests with GIT if specified in the proposal and not otherwise indicated in the SFRC.

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Date 10-9-87

Project No. G-35-643
School N/K Geo. Sci.

Includes Subproject No.(s) N/A

Project Director(s) Dr. E.M. Patterson

Sponsor Electronic Systems Division; Air Force Systems Command; Hanscom AFB, MA.

Title Optical Absorption Characteristics of Aerosol Particles

Effective Completion Date: 6/20/85 (Performance) 9/30/85 (Reports)

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- Final Invoice or Final Fiscal Report
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FORM OCA 69.265
Dr. Leonard E. Johnson  
Division of Earth Sciences  
National Science Foundation  
Washington, DC 20550

Dear Dr. Johnson,

Enclosed are two (2) copies of a paper, "Seismic structure of the Charlotte and Carolina Slate Belts of Georgia and South Carolina" by C. K. Lee and A. M. Dainty, published in Earthquake Notes 53:2, 23-38. Research for this paper was supported under NSF grant EAR 7812961. Roy Hanson was technical officer for this grant - could you please handle it for me?

Sincerely,

Anton M. Dainty

AMD: ps
Excl.
cc: OCA  /  635-643  - For you file
C. Cook
ABSTRACT: The seismic structure of the Charlotte and Carolina Slate belts was investigated by synthetic seismogram analysis. Using generalized ray theory and the Cagniard-de Hoop method, synthetic seismograms were calculated for the first few seconds of reflected and refracted P-wave arrivals and compared with observed seismograms recorded within the area being studied. A model has been obtained of the detailed velocity structure of the crust which provides a good fit for two sets of seismograms up to an epicentral distance of 210 km. This model is in agreement with reflections at 11.0 to 11.5 sec two-way vertical reflection time seen on the COCORP Georgia lines 1 and 4, and also in agreement with results from seismic refraction data assembled by Kean (1978). The model shows compressional wave velocities increasing from 5.1 km/sec to 6.0 km/sec in the upper 6 km of the crust. The average velocity of the crust is 6.0 km/sec and total crustal thickness is 33 km based on the analysis of reflections from the crust-mantle boundary, which is sharp in this area. There is a high velocity (6.6 km/sec) layer 4 km thick at the base of the crust.

INTRODUCTION

The crustal structure of the Charlotte and Carolina Slate belts in northeastern Georgia has been studied in several recent works by Dunbar (1978), Kean (1978), Long (1979), Cook et al. (1979, 1981), Kean and Long (1980) and others. There are basically two kinds of velocity structure suggested. One is a structure that has low velocity layers lying on higher velocity layers, such as Kean's (1978) model. The other has a low velocity zone beneath higher velocity layers as suggested by Long (1979), and in a different model by Cook et al. (1979, 1981). In this paper, we attempt to determine the crustal structure by calculating theoretical seismograms for proposed crustal models and comparing them with observed seismograms. According to a smoothed Bouguer Anomaly map (Long, 1974), the Charlotte and Carolina Slate belts of Georgia and South Carolina appear to be uniform in crustal thickness and composition. The synthetic seismograms calculated from homogeneous, flat-layered crustal models by the generalized ray method can thus be used to determine the crustal structure of this area.
The Charlotte and Carolina Slate belts are divisions of the crystalline Piedmont of the Appalachian orogen (Figure 1; Rodgers, 1970, p. 192-198). The rocks of the Charlotte belt are dominantly plutonic, consisting of an older matrix of granodiorite and granite gneisses along with other metamorphosed intrusive and metavolcanic rocks. The age of these rocks is not known, since they have been subject to varying degrees of metamorphism at approximately 400 m.y. B.P. The Carolina slate belt is made up of rocks of low metamorphic grade, believed to be of late Precambrian to lower Paleozoic age, and is at least 3000 meters thick. In addition to the rocks described above, there is a series of predominantly granitic plutons of ages ranging from 350 to 250 m.y. intruding both the Charlotte and the Carolina slate belts. Plutons described by Fullagar and Butler (1979) are shown in Figure 1. These plutons are post-metamorphic and cross-cut structural trends.

Figure 1. Major geological boundaries in the study area, after Williams (1978). Stippled areas are late granitic plutons discussed by Fullagar and Butler (1979).

The history and present structural position of the Charlotte and Carolina Slate belts are the subject of continuing controversy. Many workers (see, for example, Hatcher, 1972) believe that the Carolina Slate belt lies, at least in part, unconformably on top of the
SEISMIC STRUCTURE OF THE CHARLOTTE AND CAROLINA SLATE BELTS

Charlotte belt in a normal stratigraphic succession. Others (Rodgers, 1970) hold that the differences between the Charlotte belt and the Carolina Slate belt are predominantly due to differing metamorphic grade. Both hypotheses may in part be correct. Whitney et al. (1978) considered the rocks of the Carolina Slate belt as a metamorphosed primitive island arc suite laid down on oceanic crust. Long (1979) suggested on the basis of gravity that the Charlotte belt in the region of study is a continental rift zone similar to the Precambrian Keeweenawan rift of the central U.S., formed in late Precambrian or early Paleozoic.

Since the advent of the theory of plate tectonics, the Appalachian system in general and the study area in particular are believed to have been formed during the collision of North America and Africa in the Paleozoic. In the model of Hatcher (1972), the rocks of the Charlotte and Carolina Slate belts are rooted and extend to depth, although belts to the northwest have been overthrust or underthrust to a greater or lesser extent. Hatcher based this conclusion on the predominantly high angle faulting mapped in the Charlotte and Carolina Slate belts and the lack of overturned folds, as opposed to the thrust faulting and overturned folds and nappes found to the northwest. Recently, however, Cook et al. (1979, 1981) have suggested on the basis of reflection seismic profiling that the Charlotte and Carolina Slate belts, together with belts to the northwest, have been overthrust to the northwest along a master thrust at about 10 km depth by at least 200 km. The age of initiation of faulting along this proposed master thrust cannot be estimated, but faulting must have continued until at least Pennsylvanian time (~280 m.y. B.P.), since sediments in the Valley and Ridge province of this age are faulted. This model would indicate that all, or nearly all, of the surficial rocks of the Charlotte and Carolina Slate belts are allochthonous and might not be representative of rocks at depths.

DATA AND ANALYSIS METHODS

Local events and quarry blasts recorded by the Georgia Tech seismic network with epicentral distances ranging from 50 to 210 km were selected; Figure 2 shows the recording stations and the epicenters which were well located within the structure belt being studied. Large quarry blasts from a quarry in Georgia provided highly reliable data. Table 1 lists the origin times and location of the events. The seismometers used for recording stations are 1.0 Hz vertical geophones. The signal from the geophone is amplified and transmitted by phone line to a central recording site, where it is displayed on drums. In addition, events of sufficiently large amplitude are recorded on a tape system with a tape loop delay. In this study, we focused our attention on the relative amplitudes and travel times of the short-period P pulses in the first few seconds of the seismograms.

For the calculation of synthetic seismograms, the Cagniard-de Hoop method and the generalized ray method were applied. Only primary P-wave arrivals were computed and approximations introduced by Wiggins and Helmberger (1974) for increasing computing efficiency were used. Unimportant rays with small amplitudes can be cut off by setting a ray selection window. The window is such that when the critical reflection
Figure 2. Location map of seismic events and recording stations.

Table 1. Seismic events used in this study

<table>
<thead>
<tr>
<th>Event No.</th>
<th>Date</th>
<th>Time (U.T.)</th>
<th>Latitude</th>
<th>Longitude</th>
<th>Depth* (km)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>4/22/78</td>
<td>06:36:23.0</td>
<td>34.3730°N</td>
<td>81.2923°W</td>
<td>1.0</td>
<td>Earthquake</td>
</tr>
<tr>
<td>2</td>
<td>8/19/78</td>
<td>16:09:49.2</td>
<td>33.5126°N</td>
<td>82.6073°W</td>
<td>0.0</td>
<td>Blast</td>
</tr>
<tr>
<td>3</td>
<td>8/19/78</td>
<td>19:08:23.2</td>
<td>33.5126°N</td>
<td>82.6073°W</td>
<td>0.0</td>
<td>Blast</td>
</tr>
<tr>
<td>4</td>
<td>8/27/78</td>
<td>10:22:08.1</td>
<td>34.0833°N</td>
<td>81.4333°W</td>
<td>1.0</td>
<td>Earthquake</td>
</tr>
<tr>
<td>5</td>
<td>9/7/78</td>
<td>22:21:40.4</td>
<td>33.5126°N</td>
<td>82.6073°W</td>
<td>0.0</td>
<td>Blast</td>
</tr>
<tr>
<td>6**</td>
<td>9/7/78</td>
<td>22:53:17.1</td>
<td>32.9973°N</td>
<td>80.0755°W</td>
<td>1.0</td>
<td>Earthquake</td>
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<td>7</td>
<td>10/4/78</td>
<td>21:36:35.7</td>
<td>33.5126°N</td>
<td>82.6073°W</td>
<td>0.0</td>
<td>Blast</td>
</tr>
<tr>
<td>8</td>
<td>1/19/79</td>
<td>08:55:35.5</td>
<td>34.7141°N</td>
<td>82.9086°W</td>
<td>1.0</td>
<td>Earthquake</td>
</tr>
<tr>
<td>9</td>
<td>2/1/79</td>
<td>01:25:47.6</td>
<td>34.5493°N</td>
<td>81.4897°W</td>
<td>1.0</td>
<td>Earthquake</td>
</tr>
<tr>
<td>10</td>
<td>8/7/79</td>
<td>19:32:15.8</td>
<td>34.3005°N</td>
<td>81.2969°W</td>
<td>1.0</td>
<td>Earthquake</td>
</tr>
</tbody>
</table>

*Depth was constrained.
**Not used in this study.
distance falls within a range $\Delta - \Delta_1 - K\Delta_2$ and $\Delta - \Delta_1 + K\Delta_2$, then the ray is included, where $\Delta$ is the epicentral distance, $\Delta_1$ and $\Delta_2$ are the limits for selecting rays and $K$ is a weight that is proportional to the ratio of the velocity jump across the interface to the median velocity jump of the model. Stairstep approximations for continuous crustal models were made in the computation and also an earth flattening transformation was applied. Q effects were ignored since we only calculate the first few seconds of P pulses, the relative effects within a seismogram should be nearly constant, and the interpretation is primarily based on relative amplitudes and travel times within a seismogram. Johnston (1980) proposed Q values for the upper crust of the Piedmont Province of 900±100 for 10-35 Hz P-waves. For 10 Hz P-waves with a relative travel time difference of 5 sec, the relative attenuation is approximately $\exp[-0.15] = 0.86$. Thus, the error in amplitude expected from ignoring Q is of the order of ±10%, and is less than or comparable to errors from other sources.

The source wavelet (Figure 3) used in calculating synthetic seismograms was chosen to have about the same length and dominant frequency as the major phases in the observed seismograms. This wavelet should allow amplitude and travel time comparison, but not waveshape comparisons. A 10 Hz low pass filter was used for all the observed seismograms to reduce noise and lessen the effects of differential attenuation, as discussed earlier. The observed seismograms were played back from recording tapes at a speed of 25 mm/sec.

Two kinds of velocity-depth structures have been suggested for the reference area of the Charlotte and Carolina Slate belts (Figure 4). One can be represented by a model proposed by Kean (1978) using seismic refraction data, the other can be represented by a model which is an extension of Dunbar's (1978) model such that the travel times are

![Figure 3. The source function used for seismogram synthesis.](image-url)
Figure 4. Starting models based on Kean (1978) and Dunbar (1978). Dunbar's model has been added to below 12 km depth to give travel times in accord with those of Kean (1978).

consistent with Kean (1978). Kean's model is based solely on travel time measurements interpreted using standard refraction techniques. As is usual with such methods, there are no velocity inversions with depth. Kean did, however, consider the possibility of a hidden high velocity layer at the base of the crust, although he was not able to find any seismic evidence for such a layer. Dunbar suggested that a low velocity zone exists at shallow (5-10 km) depths in the crust, partly on the basis of geology.

To test these models and derive a better model, comparisons were made between observed seismograms at various distances and synthetic seismograms computed for the same distance and focal depth. In making the comparisons three criteria were used: namely, the relative amplitude of different arrivals on a given seismogram, the relative timing of different arrivals on a given seismogram, and the absolute arrival time of different arrivals. The amplitude of the synthetic seismogram was adjusted so that the largest amplitude on the synthetic seismogram was commensurate with the largest amplitude on the observed seismogram. Particular attention was placed on reflection from interfaces at near-critical angles, and accordingly seismograms in the distance range 50-150 km were weighted more heavily than others, since this is the distance range that critical angle reflections from crustal
interfaces are expected. In addition, it can be seen from the comparisons presented that the synthetic seismograms are much simpler than the observed seismograms. We have considered this to be due to scattered "noise" in the observed seismograms since we have not been able to trace any coherent arrivals. Because of this phenomenon, small arrivals on the synthetic seismograms were not heavily weighted if they were below the signal to noise level. In addition, energy arriving after the first few seconds could be due to arrivals not considered in the calculation. In general, it may be said that the interpretation is based on the major arrivals observed in the first few seconds of the seismogram.

Figure 5 shows a set of seismogram comparisons for Kean's model and Figure 6 shows a comparison of the same observed seismograms for Dunbar's model. From the two comparisons we observed reasonable agreement for Moho reflections in both models, which implies that the average crustal velocity and crustal thickness in each model are close to the true values. (The Moho reflections on the synthetic seismograms of Figure 5 and 6 are the latest in time for all distances except 197 km; on this seismogram the largest amplitude arrival is the Moho reflection). However, there are some differences for Moho arrivals. Dunbar's model shows delayed synthetic Moho arrivals for far distances. This is interpreted as indicating that the Moho depth is too deep and the average crustal velocity is too high in the model. Moho arrivals in Kean's model are more consistent with the observed data.

The first arrivals in the two models also show differences. First arrivals in near-distance seismograms correspond to structure at shallow depths. It is seen that Dunbar's model yields early first arrivals at near distances. The relative times and amplitudes for first arrivals and Moho arrivals also show large deviations in Dunbar's model relative to the observed seismograms. This indicates that the surface velocity in Dunbar's model is too high. Many other hypothetical models with a low velocity layer lying between high velocity layers were constructed and tested. None of these models are consistent with the observed data and none fits the data as well as Kean's model. Therefore, Kean's model is more likely to be close to the true structure. Working with Kean's model, comparisons and adjustments were repeatedly made in order to approach the true structure until a final model is obtained which best fits the observed data.

RESULTS

A model was found for the reference area (Figure 7). The model consists of a first layer 0.2 km thick with P-wave velocity 5.18 km/sec, a second layer 4.3 km thick with velocity 5.45 km/sec, a third layer 1.5 km thick with velocity 5.75 km/sec, a fourth layer 23 km thick with velocity 6.05 km/sec and a fifth layer 4 km thick with velocity 6.6 km/sec. The upper mantle velocity (taken from Kean, 1978) is 8.2 km/sec at 33 km depth. Seismogram comparisons are shown in Figures 8, 9 and 10 for this model.

The upper part of the crustal model for the reference area was determined using near-distance quarry blasts. Slight deviations of first arrivals in the 56 km seismograms exist for this model, probably
caused by inhomogeneities in the upper layers of the crust (Figure 10). This model presents averaged velocities for the upper layers and shows an increase of velocity with depth for the upper 6 km of crust. This increase is responsible for the first arrivals seen between 50 to 80 km. Based on the presence of these arrivals, it is unlikely there is a widespread velocity inversion in the upper 10 km of the crust.

Figure 5. Comparison of seismograms for Kean's model.
Figure 6. Comparison of seismograms for the depth extended Dunbar's model assuming the near-surface high velocity layer is continuous.
The middle part of the crust is rather uniform in this model. Any strong discontinuity of velocities in this part would cause high amplitude phases that are not seen on the observed seismograms. Other variations of velocity with depth in this region may be possible, however, as discussed below. At the base of the crust, a thin layer 4 km thick with velocity 6.6 km/sec was modeled. The reflections from this layer at different distances can be detected in the observed seismograms 0.5-1.0 second ahead of the Moho arrivals. The Moho depth and average crustal velocity were well determined from the travel time difference between first arrival and Moho wide angle reflections and agree with the vertical reflection time of 11.0-11.5 sec for a reflector seen on the COCORP profile in the reference area (Cook et al., 1979).

In assessing the reliability of the model, it should be borne in mind that this technique gives the greatest amount of information for regions of the crust from which strong returns of energy are seen, i.e. the top and bottom of the model. In these regions changes of ±1 km in depth and ±0.1 km/sec in velocity may be made without appreciably affecting the form of the seismograms. In the central part of the model, between 6 and 29 km depth, there are no strong velocity interfaces -- if there were, we would expect to see phases corresponding to them. Velocity variations are still possible, however, provided there are no strong interfaces and provided the average velocity is close to 6.0 km/sec, as determined from the moveout relation for Moho reflections. The upper mantle velocity of 8.2 km/sec was taken from

Figure 7. Crustal model A proposed for the reference area. 
\[ V_s = \frac{V_p}{\sqrt{3}} \]; densities estimated from \( V_p \) (densities have very little effect on the seismograms; Wiggins and Helmberger, 1974).
SEISMIC STRUCTURE OF THE CHARLOTTE AND CAROLINA SLATE BELTS

Kean (1978) and was not determined independently in this study. Overall, the model predicts that two P-wave arrivals will be seen in the distance range 50-150 km, one from the top of the crust and one from the bottom of the crust. Midcrustal arrivals are not predicted from the model and do not appear to be observed.

Figure 8. Comparison of seismograms (1) for model A.
Figure 9. Comparison of seismograms (2) for model A. Three additional seconds were reduced for each seismogram.
Figure 10. Comparison of seismograms (3) for model A.
The average crustal P-wave velocity of the preferred model is 6.02 km/sec. This indicates that in the reference area shown in Figure 2 the average composition of the crust is probably silicic. If geological theories (e.g. Hatcher, 1972) which hold that the rocks of the Charlotte and Carolina Slate belts are essentially in place and rooted at depth are correct, the surface rock types that outcrop in these belts and that would explain the observed velocities are the granodiorite and granite gneisses of the Charlotte belt, or the late granite plutons. Models which indicate that extensive amounts of mafic material are present in the body of the crust of this region (Whitney et al., 1978; Long, 1979) are not in accord with the model presented here, unless there has been subsequent alteration of the crust. This is possible in both the early Paleozoic island-arc model of Whitney et al. (1978) and the rift model of Long (1979) if the episode of late granitic plutonism examined by Fullager and Butler (1979) has affected large portions of the crust.

The second class of geological theories for this region, the master thrust hypothesis of Cook et al. (1979, 1981), consider the surface rocks of the study area allochthonous. If this is correct, the lithologies observed at the surface cannot be used to estimate the lithologies at depth. Cook et al. (1981) postulate on the basis of their master thrust interpretation and models of plate tectonic collision that the crust beneath the master thrust, at depths of 10 km and deeper, consists of sediments or metasediments of a continental slope-continental rise prism deposited offshore from the North American craton in late Precambrian-early Paleozoic time. In addition, this prism of sediments may be underlain by oceanic crust just above the Moho. The model presented here supports this model, at least in part. The average crustal velocity is appropriate for metasediments, and the thin high velocity layer at the base of the crust could be the oceanic crust underlying the sediments. However, no interface is found at the depth of the master thrust (10-15 km) as might be expected. This would indicate that the lithologies on either side of the thrust have similar P-wave velocities.

The results presented in this paper are certainly consistent with the master thrust model of Cook et al. (1981). Not all models which interpret the Charlotte and Carolina Slate belts as being rooted at depth (Hatcher, 1972) are consistent with the results of this paper, although some of these models are consistent with the P-wave velocity profile presented here.

ACKNOWLEDGMENTS

Discussions with L. T. Long, F. A. Cook, and B. Ellwood are gratefully acknowledged. This research was supported by the National Science Foundation under Grant EAR 7812961. Synthetic seismograms were calculated by a modification of program STPSYN3, written by Ralphe Wiggins.
SEISMIC STRUCTURE OF THE CHARLOTTE AND CAROLINA SLATE BELTS

REFERENCES


Quarterly Status Report
for period
1-25-82 to 4-30-82

Optical Absorption Characteristics of Aerosols
Project F-19852-82-K-0021
Electronic Systems Division
Air Force Systems Command
Hanscom AFB, MA

by

E. M. Patterson
School of Geophysical Sciences
Georgia Institute of Technology
Atlanta, GA 30332
The project "Optical Absorption Characteristics of Aerosols," Project F19628-82-K-0021, was initiated in January 1982. Purpose of this project has been the investigation of methodologies for making aerosol absorption measurements. During the first stage of the program, January 25, 1982 - April 30, 1982, the following tasks have been accomplished:

1. **Construction of an aerosol generation system**
   We have constructed an aerosol generation system to produce submicron sized aerosol particles from a variety of water or alcohol soluble materials. The aerosol system generates particles by means of a bubbler system. In this system filtered air is first humidified; the air then passes through the bubbler portion where the aerosols are generated, and an impingement section where the large particles are removed. Air from the generation system is mixed with dried filtered air to produce dry aerosol particles.

2. **Testing of aerosol generator system**
   The characteristics of our generation system have been investigated using both sodium chloride and ammonium sulfate aerosol. The size distributions of the aerosol have been measured using a PMS optical particle counter (LAS-X) that is part of our laboratory aerosol characterization equipment. Overall consistency of the size distribution determined by the optical particle counter has been checked by comparison of the mass concentrations calculated for the size distributions with the gravimetrically determined mass concentrations.
Agreement to within ~20% was found, indicating that our size characterization is adequate. These test aerosols have been collected onto filters suitable for SEM (Scanning Electron Microscopy) analysis for further characterization as to particle morphology.
Quarterly Status Report
for period
5-1-82 to 7-30-82

Optical Absorption Characteristics of Aerosols
Project F-19628-82-K-0021
Electronic Systems Division
Air Force Systems Command
Hanscom AFB, MA

by

E. M. Patterson
School of Geophysical Sciences
Georgia Institute of Technology
Atlanta, GA 30332
Quarterly Status Report
Report F19628-82-K-0021
Report #2

During the period 5/1/82 - 7/31/82 we continued testing the aerosol generation system with absorbing aerosols and have begun to make preliminary measurements of aerosol absorption. During this time one student, Mr. Bill Murphey, has been supported under this contract. His work on this project is expected to form part of a master's thesis.

The following specific tasks have been accomplished:

1. **Generation of Aerosols for Testing**
   
   One organic dye (Nigrosine) and two inks (Flowmaster and Mars drawing ink) have been used as source materials for the production of highly absorbing aerosols. Size distributions have been determined for the aerosols generated by each of these materials and particles have been collected onto Nuclepore filters for further characterization with the SEM facilities here at Georgia Tech.

2. **Preliminary Measurements of Aerosol Absorption**
   
   There are several ways of expressing the absorption of a material. Two widely used absorption parameters are the mass absorption parameter, $B_a$, and the imaginary component of the particle refractive index, $n_{IM}'$, with $m = n_{RE} - n_{IM}'$. $B_a$ is defined by the equation $B_a = \frac{\tau A}{M}$ with $\tau$ the measured optical depth, $A$ the area of the filter, and $M$ the mass of material on the filter. $n_{IM}'$ is defined by the equation $n = \frac{k \lambda}{4\pi}$ with $k$ the usual Lambert absorption coefficient and $\lambda$ the wavelength of the incident light.

   Nigrosine dye has been the first material studied, and some preliminary
measurements of absorption for this material have been completed.

We first made transmission measurements of the Nigrosine in solution to verify published data on this absorption. We found that our measurements at 573 nm agreed with the values published by the manufacturer to within experimental error. Measurements of absorption of this aerosol material by means of the diffuse transmission (filter transmission, integrating plate) method were then made. Expressed as a mass absorption coefficient, the solution transmission measurements resulted in a value of 3.8 m²/g at 633 nm while the filter transmission method resulted in a value of 3.4 m²/g. The different nature of the material on the filter or in the solution will certainly lead to some differences in the measured values, and further analyses to interpret these data are continuing.

In addition, preliminary measurements of absorption by means of the diffuse reflectance techniques have been done. These measurements show values somewhat lower than those measured by the other two techniques. Again, analysis is continuing.

3. Investigation of KBr Pellet Properties for Infrared Measurements

We have investigated a number of possibilities for an optical or electron microscopic analysis to characterize the physical properties of KBr pellets containing material whose absorption properties are to be measured. Based on discussions with Dr. Volz, the technical monitor, we will proceed with an SEM analysis using (NH₄)₂SO₄ dispersed in the KBr. As the first step in this analysis, we will chemically etch the surface of the pellet to allow SEM investigation of the interior of the pellet. We will then use the photographic and x-ray analysis capabilities of the SEM to characterize the (NH₄)₂SO₄ dispersion.
Quarterly Status Report
for period
8-1-82 to 10-31-82

Optical Absorption Characteristics of Aerosols
Project F-19628-82-K-0021
Electronic Systems Division
Air Force Systems Command
Hanscom AFB, MA

by

E. M. Patterson
School of Geophysical Sciences
Georgia Institute of Technology
Atlanta, GA 30332
Quarterly Status Report  
Project F-19618-82-K-0021  
Report #3

During the period 8/1/82 - 10/31/82 we used the aerosol generation system to generate a number of highly absorbing aerosols and have made absorption measurements with these aerosols. During this period, Mr. Bill Murphey has been supported under this contract for work that will form part of an M.S. thesis. The following specific tasks have been accomplished.

(1) **Generation of Aerosols for Testing and Characterization**

We have continued the generation and collection of aerosols for testing and characterization using Flowmaster and Mars inks. Size distributions have been determined for these aerosols by both electron microscopy and optical particle counters. A representative set of these size distributions is shown in Fig. 1. SEM photographs of the Flowmaster ink aerosols are shown in Fig. 2. The picture indicates that the particles are spheroidal with only slight deviations from sphericity. The differences in the size measurements indicate that the optical particle counter is not suitable for the primary size characterization of the particles.

(2) **Absorption Measurements**

Detailed calibration measurements of the transmission of the Flowmaster and Mars inks were made. An example of the raw data used to check the Beer's Law behavior of the Flowmaster ink is shown in Fig. 3. Each of the inks was found to be well behaved optically. Preliminary measurements of absorption by means of diffuse transmission measurements were made for each of these inks. A plot of \( \tau \) vs mass loading for the Mars ink collected onto Nuclepore filters is
shown in Fig. 4. There appears to be a linear relationship between the optical depth and the mass loading for the two inks investigated.

(3) **Investigations of KBr Pellet Properties for Infrared Measurements**

KBr pellets containing ammonium sulfate particles were etched with water and analyzed with the SEM using X-ray fluorescence. We attempted to use the energy dispersive X-ray analyzer on the SEM to map out the elemental S distribution in the pellet as a means of determining the distribution of the ammonium sulfate particles. The sensitivity of the X-ray analyzer, however, was not sufficient to map out the distribution as we had wished. We are planning microprobe measurements for increased sensitivity,
## Project F-19628-82-K-0021

3rd Quarterly Report 8-1-82 to 11-30-82

Updated Cumulative Budget Information for 3rd Report Period

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<td><strong>Total Costs</strong></td>
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Direct costs include expenditures and encumberances (as when an order is placed). Overhead is not charged when money is encumbered so amount does not match contract rate precisely. The overhead rate is 47.2% of direct costs, excluding capital equipment costs. The above budget information does not include an approximately $500 instrument repair during this period for which funds had not been paid or encumbered.
Figure 1.

VOLUME DISTRIBUTION COMPARISON BETWEEN KNOLLENBERG (PMS=CIRCLES) AND SCANNING ELECTRON MICROSCOPE (SEM=+'S), FLO-MASTER INK (NUCLEPORE FILTER)
Figure 2.
Figure 3.

FLO-MASTER: SOLN. TRANS. (573 NM)

TAU VS. CONCENTRATION  SLOPE=239.3

CONC. (G/L)
Figure 4.
Quarterly Status Report
for period
11-1-82 to 1-31-83

Optical Absorption Characteristics of Aerosols
Project F-19628-82-K-0021
Electronic Systems Division
Air Force Systems Command
Hanscom AFB, MA

by

E. M. Patterson
School of Geophysical Sciences
Georgia Institute of Technology
Atlanta, GA 30332
The period 11-1-82 to 1-31-82 was primarily devoted to absorption measurements of the highly absorbing ink and dye materials. We also began some measurements on glass with glass filter material and continued our attempts on KBr pellet characterization. The following specific tasks have been accomplished:

1) Measurements of Ink Absorption by Diffuse Transmission

We continued our diffuse transmission measurements with the ink and dye aerosols, using different filters as collection substrates. These measurements were made to investigate the different response characteristics of these filter materials. A series of such measurements were made using Nuclepore filters, glass fibre filters, and Millipore filters as examples of three types of filters in common use: capillary pore, tortuous pore, and fiber mats. Examples of tau vs mass loading plots for each of these filter types are shown in Figs. 1-4. These plots do not indicate major differences in filter response for these filters for the ink aerosol. This data may be contrasted with previous data on soots and natural aerosols which do indicate some significant differences in filter response. The reasons for these differences are presently being investigated.

2) Measurements of Diffuse Reflectance

Preliminary diffuse reflectance measurements were made for the inks and the glass samples. Measurements for the flowmaster ink are shown in Fig. 5. The solution transmission value as a function of wavelength is shown as the heavy solid line. Diffuse reflectance measurements for several concentrations
are shown in the figure. The low concentration diffuse reflectance measurements (.1 and .4 mg mass loading) are self consistent and in fairly close agreement over much of the wavelength range with the transmission measurements. Further measurements will be continued.

(3) KBr Pellet Characterization

We attempted to make microprobe measurements on the KBr samples. We have had equipment problems, however, and have not yet been able to make these measurements.
Figure 1.

FLO-MASTER (NUCLEPORE)

SLOPE=1.91
Figure 2.

FLO-MASTER (GLASS FIBER)

SLOPE = 1.74
Figure 3.

FLO-MASTER (MILLIPORE .45 MICRON)

SLOPE = 1.73
Figure 4.

FLO-MASTER (MILLIPORE 1.2 MICRONS)

SLOPE = 1.69

TAU

MASS

Figure 4.
FLO-MASTER INK: COMPARISON BETWEEN DIFFUSE REFLECTANCE (+'S, TRIANGLES, CIRCLES, DIAMONDS), SOLUTION TRANS. (X'S), AND LASER TRANS. VALUE (LONE CIRCLE)

Figure 5.
Quarterly Status Report
for period
2-01-83 - 4-30-83

Optical Absorption Characteristics of Aerosols
Project F-19728-82-K-0021
Electronic Systems Division
Air Force Systems Command
Hanscom AFB, MA

by

E. M. Patterson
School of Geophysical Sciences
Georgia Institute of Technology
Atlanta, GA 30332
The period 2-1-83 to 4-30-83 was primarily devoted to measurements for the absorption of glass filter material and analysis of the absorption measurements of highly absorbing inks and dyes. The following specific tasks have been accomplished.

(1) **Measurements of the Absorption of Filter Glass**

We have analyzed some optical filters. These optical filters were chosen for the analysis in the expectation that the glass particles would not interact with the BaSO₄ used as a standard in the reflectance analysis. In addition, the absorption of the glass could be measured directly without the necessity of producing a solution of the absorbing material in a solvent.

For these measurements, our procedures involved direct transmission measurements of the filters from which specific absorption values as a function of wavelength were determined. The material was then ground, and diffuse reflectance measurements of the absorption of this powder were made. These measurements have been made for two different Schott glass filters, UG8 and BG38.

Transmission measurements for the filters are shown in Fig. 1(BG38) and Fig. 2(UG8) as dotted lines. These measured values of the specific absorption $B_a$ are quite similar to those determined on the basis of the published filter absorption data (shown as the solid line in Fig. 1 and Fig. 2). The comparison of the $B_a$ values determined by the transmission and the reflectance
methods is shown in Fig. 3(BG38) and Fig. 4 (UG8). The transmission values are shown as the heavy solid line and the reflectance values are shown by the other lines in each figure. For both of the filter glasses, the reflectance measurements show absorption values significantly higher than those measured by transmission, with a different wavelength dependence. The data shown in Fig. 3 suggest that there is no significant effect of differing concentration on our reflectance measurements. The variation of the measured absorption with variation in grinding time for the reflectance data shown in Fig. 4 does indicate that the process of grinding the glass does affect the absorption properties of the glass. Further measurements are planned to try to determine the magnitude of any such grinding effects.

(2) **Aerosol Sample Characterization**

Scanning electron microscopic analyses of several aerosol samples were made using Georgia Tech facilities. These measurements were made to characterize laboratory samples of aerosol collected for measurements of optical properties. This characterization included aerosol size, shape, morphology, and collection characteristics.
GLASS TRANSMISSION VALUES FOR BG38 COLOR FILTER GLASS: COMPARISON BETWEEN OBSERVED VALUES (TRIANGLES) AND CALCULATED VALUES (CIRCLES)

Fig. 1
GLASS TRANSMISSION VALUES FOR UG3 COLOR FILTER
GLASS: COMPARISON BETWEEN OBSERVED VALUES (CIRCLES) AND CALCULATED VALUES (TRIANGLES)

Fig. 2
GLASS TRANSMISSION VALUES FOR BG38 COLOR FILTER
GLASS: COMPARISON BETWEEN OBSERVED VALUES OF SOLID
GLASS (DARK LINE) AND GRINDED GLASS (+S.TRI.CIR.DIA)

Fig. 3
GLASS TRANSMISSION VALUES FOR UG3 COLOR FILTER GELASS: COMPARISON BETWEEN OBSERVED VALUES OF SOLID GLASS (DARK LINE) AND GRINDDED GLASS (+S, TRI, CIR, DIA)
Quarterly Status Report
for period
5-1-83 to 7/31/83

Optical Absorption Characteristics of Aerosols
Project F-19628-82-K-0021
Electronic Systems Division
Air Force Systems Command
Hanscom AFB, MA

by

E. M. Patterson
School of Geophysical Sciences
Georgia Institute of Technology
Atlanta, GA 30332
The period 5-01-83 to 7-31-83 involved some differences in direction for the project. There was a continuation of our analysis of the transmission and reflectance of aerosol samples with visible light and samples were collected for analysis of visible and infrared wavelength absorption properties. A major portion of our effort was devoted to an attempt to estimate some of the infrared optical properties of the ash from El Chichón volcano that involved comparisons of different infrared measurement techniques.
El Chichón Volcano

One of the major events of the past few months was the May 1983 eruption of the Chichon volcano in Chiapas, Mexico. This has led to a very large perturbation in stratospheric aerosol optical depth. Various analyses of measurements of stratospheric temperatures after the eruption have shown that there was a significant heating of the stratosphere by the Chichon aerosol cloud. Analysis of visible wavelength absorption data has indicated that heating by the direct absorption of solar radiation by the aerosol cloud is not sufficient to account for the observed stratospheric heating and that infrared effects may well be the dominant effect. The initially large sizes for the silicate ash component of the aerosol suggests that this silicate ash may have significant infrared effects. We have not ourselves made any infrared measurements of the material, although some material collected at the surface is available for analysis. We have, however, re-examined some earlier infrared measurements of ours and others in order to make a preliminary estimate of possible infrared effects.

Infrared Measurements of Volcanic Ash

The basic measurement set is one reported by our group several years ago (Patterson, 1975). This data set consists of measurements of infrared absorption properties between 1 and 15 \( \mu \text{m} \) for several samples of volcanic ash. These measurements were made using the potassium bromide pellet technique with interpretation of the transmission data in terms of a specific absorption coefficients \( k_p \), from which imaginary components of the index of refraction, \( n_{\text{IM}} \), were calculated as a function of wavelength. Bulk material was assumed and no corrections for the actual aerosol nature of the material in the pellet were made.

These measurements showed that the volcanic ash showed relatively small variation in these samples which ranged from basaltic to rhyolitic in composition. The
average \( n_{\text{IM}} \) curve for this set of samples is shown as Fig. 1 of this progress report. The error bars at the various wavelengths show the standard deviation in the absorption values measured at the given wavelengths for these samples.

Our further analysis as part of this study of absorption measurements have involved an investigation of the variations in the absorption with variation in composition among this set of samples, a correction of the absorption in terms of a correction factor discussed by Volz, a comparison with transmission measurements of thin slabs of uniform material (Pollack et al., 1973), and a prediction of results of Chichón.

The variation in \( n_{\text{IM}} \) at 9.5 \( \mu \text{m} \) with variation in the \( \text{SiO}_2 \) composition of the ash is shown in Fig. 2. The circles (o) represent the 9.5 \( \mu \text{m} \) values from the data set shown in Fig. 1. The diamond (◊) is a value taken from the data of Volz for volcanic material. The Volz value was characteristic of Hawaiian volcanos. Overall composition data for Hawaiian volcanos was used to plot this absorption value at 49% on the \( \text{SiO}_2 \) composition axis. It is apparent from the figure that the Volz data are consistent with the other data set.

In general, it appears that increases in \( \text{SiO}_2 \) component are associated with increases in \( n_{\text{IM}} \).

**Corrections to Infrared Data**

An approximate fit of the absorption data to the \( \text{SiO}_2 \) percent composition is shown as the solid line in Fig. 3. Volz has discussed correction factors that may be applied to infrared data to account for the fact that the pellets consist of particles embedded in a substrate rather than a non scattering bulk material. The approximate correction factor of 1.6 discussed by Volz may be applied to this volcanic data. This "corrected" data is shown by the dashed line in Fig. 3.
Comparison with Reflectance Data

This corrected data may be compared with other IR data measured by Pollack et al. The Pollack data were determined using a dispersion analysis of reflectance data from highly polished crystalline sections of their volcanic material. The Pollack data consists of only 5 data points, one group of these at about 53% SiO₂ and a group of two at ~73% SiO₂ composition. These Pollack data are systematically higher than our KBr pellet data, but the trend of increasing absorption with increasing SiO₂ percent composition is quite similar.

If there is indeed a relationship between the SiO₂ percent composition and the absorption, as implied by our data, then we can infer that these differences between our data and the Pollack data are primarily due to systematic differences in the measurement techniques. The overall differences, however, are within a factor of 2.

Because of the uncertainties in the KBr pellet technique, at this time for these particular data sets, we feel that the crystallographic data is a more accurate representation of the actual absorption that the pellet data.

Infrared Value of n_{IM} for Chichon Ash

We may use this comparison to infer an n_{IM} value at 9.5 μm for the Chichon ash. Chemical measurements show that the SiO₂ component represents ~59% of the ash. Based on this compositional data, making use of the Pollack absorption data, we may infer that the n_{IM} is ~1.2 at 9.5 μm, a value considerably higher than might have been inferred from our previous data.

This comparison must be regarded as preliminary at this time, but further work applying these ideas is planned.
References


AVERAGE IMAGINARY INDEX OF REFRACTION FOR ALL VOLCANIC ASH SAMPLES

Fig. 1
Fig. 2

VOLCANIC ABSORPTION COMPARISON

IMAGINARY INDEX OF REFRACTION

SILICON DIOXIDE COMPONENT (PERCENT)
VOLCANIC ABSORPTION COMPARISON

IMAGINARY INDEX OF REFRACTION

SILICON DIOXIDE COMPONENT (PERCENT)

Fig. 3
OPTICAL ABSORPTION CHARACTERISTICS OF AEROSOLS

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11 September 1985

Final Report
26 January 1982–30 June 1985

APPROVED FOR PUBLIC RELEASE; DISTRIBUTION UNLIMITED

AIR FORCE GEOPHYSICS LABORATORY
AIR FORCE SYSTEM COMMAND
UNITED STATES AIR FORCE
HANSCOM AIR FORCE BASE, MASSACHUSETTS 01731
The absorption of well documented, highly absorbing submicron aerosols has been measured by means of diffuse reflectance and diffuse transmission techniques. The intent of these measurements was to compare the different techniques and to provide calibration of these absorption measurements in terms of aqueous transmission measurements.

These measurements indicate that there is a consistency among the absorption techniques that the differences between absorption in an aqueous and in an air suspension need to be considered in calibrations of absorption techniques, and that the diffuse transmission and diffuse reflectance measurements using the dilution technique appear to yield consistent results.
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INTRODUCTION

The absorption of visible and near visible radiation by the atmospheric aerosol is often a significant component of the aerosol radiative effects. This absorption is important because of its direct effect on visibility reduction and on radiative transfer in the atmosphere (see e.g. Groblicki et al, 1984), as well as its possible effect on radiative transfer in optically thick cloud layers (Patterson et al, 1984). The possible climatic effects of the aerosol, for example, will be determined in large part by the ratio of absorption to scattering in the aerosol. In addition, the interpretation of remotely sensed data and the proper consideration of atmospheric effects on this data requires knowledge of the absorption properties as well as the scattering properties of the aerosol.

The aerosol absorption can have a very wide range of variation. Expressed as a percentage of the total light extinction by an aerosol, the absorption at visible wavelengths can range from much less than 1% of the extinction in a white, low absorption material such as ammonium sulfate or ammonium nitrate to more than 90% of the total extinction in highly absorbing soots and smoke emissions. The wide range of the measured absorption under different ambient conditions was shown by Gerber (1981).

For highly absorbing materials, in particular, an accurate
assessment of the aerosol radiative effects requires accurate determinations of the aerosol absorption. There appear, however, to be some significant systematic differences in the results obtained using different experimental techniques. These differences were shown in a recent set of absorption measurement intercomparisons discussed by Gerber and Hindman (1981). There was a range of roughly a factor of two among the various absorption measurements for both a soot aerosol and a methylene blue aerosol measured in the intercomparison.

While these intercomparisons provided valuable insights into the limitations of the different absorption measurement methodologies, no attempt was made in that study to choose any one of the methodologies as a standard against which the others could be judged. There was also no attempt to make calibration measurements with aerosols whose absorption was determined independently of the intercomparison. And, in general, there have been relatively few calibrations for the commonly used absorption techniques using materials for which the bulk optical constants are known independently.

We have made a series of absorption measurements of laboratory aerosols that are an extension of the work presented in Gerber and Hindman. The goal of the measurements discussed here was twofold: first, to make further comparisons of several methodologies for measuring aerosol absorption and second, to go beyond a simple comparison of methods by providing a calibration for the measured
aerosol absorption in terms of standard, well determined measurements. The experimental procedures included laboratory generation of aerosols from several different materials, measurement of aerosol absorption by two diffuse reflectance techniques and one diffuse transmission technique, and measurement of the absorption of an aqueous suspension or solution of each material. The measurements emphasized a study of an aqueous suspension of carbon black (Mars ink) as a reproducible analog of the soot which is believed to be responsible for much tropospheric absorption. As a stable suspension with well defined and documented physical properties it is well suited for our characterization measurements.

Variations in the individual techniques were investigated; in addition, absorption measurement comparisons were made for other aerosols that were not as suitable for such extended laboratory studies.

In this report, several comparisons of results of different methods of measuring absorption for a variety of materials are presented; and some of the limitations of the different methods are discussed. The measured aerosol absorption is discussed in relation to the absorption of an aqueous suspension which provides a calibration for the aerosol measurements. The Mars ink results are discussed in considerable detail.
The results of aerosol absorption measurements can be reported in terms of absorption parameters that are characteristic of the aerosol suspension or of the bulk material making up the aerosol. For a bulk non-scattering material the absorption may be described in terms of the Lambert absorption coefficient \( k \), with \( k \) defined by the equation

\[
k = -\frac{1}{x} \ln \left( \frac{I}{I_0} \right)
\]  

with \( x \) the path length through the absorbing material, \( I_0 \) the incident intensity, and \( I \) the intensity of the transmitted light. The turbidity \( \delta \) is a measure of the total light reduction by the absorbing material and is given by the equation

\[
\delta = -\ln \left( \frac{I}{I_0} \right)
\]  

\( \delta \) and \( k \), then, are related by the equation

\[
k = \frac{\delta}{x}
\]  

For the usual laboratory transmission spectroscopy measurements, the value of \( x \), the path length through the sample, is not always a readily measurable quantity. Another absorption parameter, the specific absorption \( B_a \), may be defined in terms of the directly
measured $\delta$ and the ratio $M/A$, the mass of absorbing material $M$ per unit area $A$ illuminated by the spectrometer beam. As defined, a determination of $B_a$ does not require a knowledge of the density $\rho$ of the absorbing aerosol.

The equation for $B_a$ can be written in terms of these directly measured quantities as

$$B_a = \frac{\delta A}{M} \tag{4}$$

$B_a$ is then related to the Lambert absorption coefficient, $k$, by the equation

$$B_a = \frac{k}{\rho} \tag{5}$$

Another widely used absorption parameter is the imaginary part of the complex index of refraction, $n_2$, which is used in Mie calculations. For a bulk non-scattering material, $n_2$ is related to the specific absorption by the equation

$$n_2 = \frac{B_a \rho \lambda}{4\pi} \tag{6}$$

with $\lambda$ the wavelength of the incident light.

When the absorbing material consists of discrete absorbing and scattering particles imbedded in a material with a differing refractive index, the above relations must be modified to take the effects of scattering into account. The aerosol absorption may be charac-
terized by a volume absorption coefficient $\sigma_a$, which is defined analogously to the extinction coefficient $\sigma_e$ and the scattering coefficient $\sigma_s$; it is defined as the total energy removed by absorption from the incident beam per unit path length by the aerosol particles. For the aerosol suspension,

$$\sigma_a + \sigma_s = \sigma_e. \quad (7)$$

A specific absorption may be defined for the aerosol in an analogous fashion to the definitions in Eq (4) and Eq (5) as the ratio of the volume absorption coefficient to the mass concentration of the aerosol, $M_v$, according to the equation

$$B_a = \frac{\sigma_a}{M_v}. \quad (8)$$

The equivalence between the definitions in Eq (4) and Eq (8) may be seen by considering the mass of aerosol material in a volume that is defined by a cross sectional area $A$ and a length $x$. If both the numerator and the denominator of Eq (8) are multiplied by the length $x$ then the numerator can be written as $\delta_a$, the turbidity due to absorption; and the denominator is the mass per unit area responsible

6
for producing the $\delta_a$. It must be remembered, of course, that the scattering is responsible for a significant portion of the extinction and that $B_a$ is defined in terms of the absorption only. Laboratory measurements of aerosol absorption generally involve the collection of aerosol material; and the assumption is made that the absorption is not changed by the process of collection. $B_a$ for the aerosol can then be inferred from the laboratory measurements if the effects of aerosol scatter can be accounted for in an appropriate manner.

For an aerosol, the relationship between $B_a$ and $n_2$ depends in part on the real component of the refractive index, $n_1$, of the discrete particles relative to that of the surrounding medium; and Eq (6) must be modified. For an aerosol consisting of particles that are small relative to the wavelength of the incident light and that have $n_1$ values between 1.25 and 1.75 and $n_2$ values of less than 1, Hänel and Dlugi (1977) have used the equation

$$n_2 = \frac{B_a \rho \lambda}{4 \pi} \frac{(n_1^2 + 2)}{3}$$

which is derived from an expression of Penndorf (1962) to approximate the relation between $B_a$ and $n_2$.

Bohren and Huffman (1983) present a slightly different equation relating $n_2$ to $B_a$.
\[ n_2 = \frac{B_a \rho \lambda}{4 \pi} \left[ \frac{(n_1^2 + 2)^2}{9 n_1} \right] \]  \hspace{1cm} (10)

Both equations have the same range of validity; within their range of validity, the two equations differ by \( \approx 5\% \). Both are strictly applicable only in the Rayleigh (small particle) limit; for somewhat larger particles, the relationship between \( n_2 \) and \( B_a \) is more complex and is dependent on particle size as well as refractive index value. For particles that range in size up to size parameter \( x = 1 \) (with \( x \) defined as the ratio \( x = 2\pi r/\lambda \), with \( r \) the particle radius) the approximate relationship will be accurate to within about 40\% compared with exact Mie calculations.

An example of the relation between the approximations of equations (9) and (10) and the exact Mie calculations, expressed in terms of the absorption efficiency \( Q_a \) is shown in Fig 1. The solid line is a plot of the absorption efficiency determined by exact Mie calculations. The dashed line is a plot of the absorption efficiency to be expected using Equation (10), assuming the given refractive index. It is apparent from the curve that the use of Equation (10) to determine a value of \( n_2 \) from the measured \( B_a \) will lead to an overestimation of \( n_2 \) for size parameters near 1. For aerosols in this size range a more accurate inference of \( n_2 \) from the \( B_a \) data can be made using the appropriate Mie calculations.

Because \( B_a \) depends on \( n_1 \) as well as on \( n_2 \), the \( B_a \) value mea-
sured for a highly absorbing particle will depend on the value of \( n_1 \) relative to the surrounding medium. As Bohren and Huffman (1983) have discussed, the measured absorption for an aqueous suspension of small highly absorbing particles will be expected to be greater than that of an air suspension of the same particles. Comparisons between measurements of aqueous and air suspensions must take these differences into account.

EXPERIMENTAL METHODS

Materials Studied

In this study, aerosols were generated using Staedtler Mars 745 black drawing ink, Flo-Master ink, and nigrosine dye. The Mars ink consists of gas-soot (carbon black) particles with a dispersing agent in an aqueous suspension. The carbon black particles have an approximately log-normal size distribution with a mass mean diameter of about 0.09 and a \( \sigma \) for the distribution of 1.6 (Hogan et al, 1985). The Mars ink consists of \( \approx 0.22 \) g of solid material per ml of ink; the density of the solid material in the ink is \( \approx 1.8 \) g/cm\(^3\).

The Flo-Master ink, unlike the Mars ink, consists entirely of material that is soluble in organic solvents. The nigrosine dye is a biological stain that is available from Eastman Organic chemicals. The solid nigrosine dye has a density \( \rho \) of 1.4 g/cm\(^3\). The nigrosine dye solution is stable, with stable absorption properties. Its behavior in solution may be referenced against standard absorption
data determined by the dye manufacturer. These materials were chosen because of their high absorption, the ease of generation of aerosols using these materials, and the ability to characterize the generated aerosol particles. Methylene Blue, the aerosol used in the earlier absorption intercomparison, was not used because our experience subsequent to the earlier study showed that this material was not stable and that its optical properties changed with time.

In addition to these laboratory aerosols, absorption measurements were made for volcanic ash aerosols and for smoke emissions from laboratory fires using diffuse transmission and diffuse reflectance techniques.

**Aerosol Generation**

The aerosols were generated using an atomizer system in which humidified compressed air was bubbled through a suspension (or solution) of the ink. The nigrosine dye and the Mars ink were diluted with water; for the flowmaster ink, an organic solvent was used for the dilution. The resulting aerosol was passed through an impaction system to remove the larger particles before drying. The aerosol generation system produced relatively large quantities of aerosol particles with stable size distributions.

Sizes of the generated aerosols were measured from scanning electron micrographs; size distributions were also measured using a single particle optical counter. An example of the size distribution measured for the Flo-Master ink is shown in Fig. 2. The curves
show data determined from the micrographs as well as data determined from the optical particle counter. There is rough agreement between the curves, even though no explicit calibration for the absorbing aerosol was made. We can see from the curves that the mass of the aerosol is concentrated in the sizes between 0.1 and 1 μm. The sizes of the nigrosine particles were slightly smaller than the sizes of the Flo-Master particles shown in Fig. 2. The size distributions for the Flo-Master ink and the nigrosine dye depended on the operating characteristics of the generation system and on the initial concentration of the ink. Since the Mars ink consisted of a suspension of solid particles the size distribution of the aerosol was more strongly influenced by the original size distribution of the carbon black particles. At the dilutions used in the aerosol generation system for this study, however, some of the generated particles contained more than one of the carbon black particles and there was some agglomeration on the filters.

Absorption Measurements

Absorption measurements were made using a diffuse transmission technique and two diffuse reflectance techniques; absorption measurements were also made using solution transmission techniques.

The diffuse transmission (DT) method of measuring aerosol absorption, which has been described by Patterson and Marshall (1982), is a variation of the integrating plate method described by Lin et al (1973) and the laser transmission technique of Rosen et al
(1978). Essentially, the technique involves a measurement of the intensity of laser light that is diffusely transmitted by a loaded aerosol filter relative to that transmitted by a blank filter and the calculation of a turbidity $\delta$ from these relative intensities. The filter is backed with a plastic diffuser to produce diffusely transmitted light whose angular distribution of intensity has a $\cos \theta$ dependence, where $\theta$ is the angle from the normal to the diffuser surface. With this diffuser, the angular dependence of the transmitted light is the same, whether the aerosol is present or not. The measured intensity does not depend on the details of the forward scattering by the aerosol sample, and the reduction in measured intensity is due only to the light absorbed by the sample and to that backscattered by the sample. The backscattered fraction of light, then, must be much smaller than the absorbed fraction. For most aerosols, the backscatter fraction is less than 10% of the total scattering and this requirement will be met except in cases in which the absorption is very low. This requirement is met for the highly absorbing samples considered in this study.

In this DT method, the $\sigma_a$ values are determined from the turbidity $\delta$ of the filter, the area of the filter and the air flow through the filter during the sample collection. $B_a$ values are determined from the above data and the mass of material on the filter. It has been shown that the response of these transmission methods can be dependent on filter type and orientation (Patterson and Marshall, 1982). Various studies (see for example Patterson and Mar-
shall) have shown that the use of a Nuclepore filter with the aerosol sample oriented toward the incident laser beam will minimize any enhancement due to filter effects. This study, then, has emphasized measurements with samples collected onto Nuclepore filters.

The diffuse reflectance techniques make use of the fact that the total diffuse reflectance of a powder surface is determined by the absorption and scattering properties of the powder as well as the thickness of the layer. For a layer that is thick enough so that no light is transmitted, the Kubelka-Munk theory, which is a two-stream radiative transfer model, relates the reflectance to the ratio of the absorption to the scattering. The Kubelka-Munk theory has been shown to be an accurate description of the reflectance when there is no specular component of the reflectance and when the absorption is small relative to the scattering.

Two diffuse reflectance techniques were studied. In one, the dilution method (DR), the aerosol material is mixed with a highly reflecting barium sulfate powder and the total diffuse reflectance of the mixture is measured using an integrating sphere. The reflectance of the mixture is, in general, less than the reflectance of the pure barium sulfate. The dilution is great enough so that the scattering properties of the mixture are the previously measured scattering properties of the barium sulfate, and so the decrease in reflectance due to the addition of the aerosol is attributed solely to aerosol absorption. The absorption of the aerosol can then be determined from the known concentration of the aerosol in the mix-
ture and the absorption of the mixture with the application of an appropriate mixing rule. The usual assumption is that volume average mixing rules apply. A more complete description is given in Patterson et al (1977) and Patterson and Marshall (1982).

According to the Kubelka-Munk theory, for finely divided powders, the DR technique provides a direct measure of the $B_a$ for the bulk material; $k$ can be determined if the density of the aerosol material is known. Under the assumptions of the Kubelka-Munk theory, the imaginary component of the refractive index is determined using Equation (6).

The other diffuse reflectance method studied is a filter reflectance (FR) method that was originally described by Lindberg and Snyder (1973). This reflectance method also uses the Kubelka-Munk formalism. In the FR method the reflectances of both a loaded Nuclepore filter and a blank filter are measured with a dark and with a white background. These four reflectance values are interpreted to determine the Kubelka-Munk scattering and absorption for the aerosol, as well as the bulk absorption $k$ and the imaginary component of the refractive index $n_2$.

The solution transmission measurements used dilutions of the inks and dye in standard spectrometer cells. The absorption of the material was inferred from the measured turbidities and the known concentrations of the materials in the cells using standard procedures. It was expected that these materials should follow Beers law, with the turbidity proportional to the concentration, for low
concentrations; for higher concentrations, deviations from such linearity allow the estimation of the effects of scattering. For low concentrations, the absorbing materials followed Beers law, as expected. In one of the cases, that of the Mars ink, deviations from linearity were used to infer a contribution of scattering to the turbidity.

The DT and the FR measurements used the aerosols generated by the generation system. The DR measurements were made using an evaporative residue for the bulk material. For dilute material in which there is a specific residual particle size such as the Mars ink, this should cause no problems. For the other inks and dyes, this difference in technique must be considered in the analysis of the comparisons.

ABSORPTION RESULTS AND COMPARISONS

Comparisons of absorption methodologies were made for the DR, the FR, and the DT techniques for the laboratory generated materials as well as for the additional materials considered. As discussed above, an emphasis was placed on the Mars ink measurements as an analog for atmospheric soot. The Mars ink carbon black suspension, the other laboratory aerosols, and the additional materials are discussed separately below.
Absorption of a Carbon Black Suspension (Mars Ink)

Several series of measurements were made using the Mars ink carbon black suspension. Aqueous transmission measurements were made using a 1.0 cm path length cell with different dilutions of the Mars ink. Turbidity values were calculated from the data; a set of these turbidity values vs mass concentration of the solid ink material is shown in Fig. 3.

Since the data are for transmission through an aqueous suspension of the particles, the extinction is due to both scattering and absorption. As a result the plot of turbidity vs mass concentration is not a straight line at the higher turbidities and there are deviations from Beers law. The deviations from Beer's law for the different ink dilutions were used to infer in an approximate way the relative importance of scattering in producing the extinction. This was done by calculating the best fit line of turbidity vs mass concentration for the low concentration data that is shown in Fig. 3 and considering that this would be the expected turbidity in the absence of multiple scattering. A radiative transfer routine was used to calculate the expected transmission for differing values of \( \omega \), the ratio of the scattering to the extinction, for expected turbidity values of 3 and 4. The inferred \( \omega \) was that which best reproduced the observed transmission values. Using this procedure, we inferred an \( \omega \) of 0.15, which is in general agreement with the data of Hogan (1984), who also inferred that scattering was only a small portion of the extinction of the aqueous suspension.
Use of this value of $\omega$, together with the extinction values inferred from the low concentration transmission data and the data on the mass concentration in the suspension, allows us to determine a value of $B_a$ of 3.5 m$^2$/g at $\lambda=633$ nm for the aqueous suspension of the solid material. Following the discussion in Bohren and Huffman (1983), this measured value of $B_a$ for the aqueous suspension will not be the $B_a$ value expected for an aerosol suspension. If a value of 1.7 is assumed for the real component of the refractive index in air, then the aqueous data imply a $B_a$ value of 2.6 m$^2$/g for the particles as an air suspension.

It should be pointed out that this difference in absorption will affect measurements such as those of Heintzenberg (1982) in which the absorption of a liquid suspension is measured. His reported value of 9.68 m$^2$/g for $B_a$ for an aqueous suspension will actually correspond to a somewhat lower aerosol value.

A plot of $\delta$ determined using the diffuse transmission technique for $\lambda = 633$ nm is shown in Fig. 4. Attempts to fit the total data set to a single regression line resulted in a $y$-intercept value that was significantly greater than 0, indicating some inconsistencies in the data. Use of the low concentration turbidity data only did result in a $y$-intercept that did not differ significantly from 0 and so this low turbidity, low concentration data was used in the subsequent analysis. The filter area is 14 cm$^2$ and the low mass loading data imply a $B_a$ of 2.4 m$^2$/g.

Measurements of $B_a$ were made using the DR and the FR methods.
The DR method, which involved the dilution of the sample, resulted in a value of $B_a$ of 3.0 at 633 nm. For these highly absorbing particles, however, great care had to be taken to insure that the particles were not aggregated into much larger particles for which the effective $B_a$ would be lower than the $B_a$ for the separate particles; for those cases in which such care was not taken, the measured values were lower than our reported values by factors of 2 to 3. The FR method, by comparison, produced results that were somewhat lower than the other measured values by approximately a factor of 2.

An overall comparison of the results from the aqueous transmission measurements, the DR measurements, and the DT measurements is shown in Fig. 5. It is apparent that there is a substantial agreement among these three methods at the 633 nm wavelength as well as at the other wavelengths measured. Since the aqueous transmission measurements provide a direct measure of the absorption, the agreement between the corrected values determined from the aqueous measurements and the DT and DR measurements indicates that there are no substantial systematic errors in these aerosol measurements. There do appear to be some systematic errors in the FR measurements. These errors may be related to the high absorption of the samples, since the Kubelka-Munk theory will not produce as accurate results for high absorption powders as for low absorption powders (Kortum, 1969).

Measured $B_a$ values have been used to calculate $n_2$ values at 633 nm for each of the measurements shown in Fig. 5. $n_2$ for the DR
data was calculated using Eq (6). \( n_2 \) for the two other measurements was determined from Mie calculations of absorption for the size distribution of the Mars ink using a range of \( n_2 \) values appropriate to both air and water. The \( n_2 \) value that reproduced the measured \( B_a \) values was considered to be the best value. We would add the caveat that the actual sizes on the filters are somewhat larger than indicated due to agglomeration and that the actual value of \( n_2 \) for the Mars DT data will be somewhat less than indicated.

The results of these calculations are shown in Table 1. It is apparent that there is a substantial agreement in the \( n_2 \) values as shown in the Table. The values for \( n_2 \) that would be calculated with the use of Eq (10) are also shown for comparison.

### Nigrosine Dye Measurements

Nigrosine absorption was determined for an aqueous solution of the dye and for an aerosol consisting of the dry nigrosine material formed by the evaporation of the droplets produced by the aerosol generation system. Nigrosine absorption data are shown in Figs. 6 (aqueous solution data) and 7 (aerosol data). The aqueous data were determined by standard solution techniques with a 10 cm cell; the aerosol data were determined by the DT technique. These solution data were consistent with the manufacturer's specifications for absorption. Since the nigrosine dye forms an aqueous solution rather than a suspension, the effects attributed to discrete particles were not observed in the aqueous data.
The plot of turbidity vs mass concentration for the nigrosine shows no deviations from linearity that indicate a significant contribution of scattering to the extinction; consequently no adjustments to the aqueous data to take account of scatter were made. The data imply a $B_a$ of $3.6\text{m}^2/\text{g}$ at 633 nm for the Nigrosine in an aqueous solution. The diffuse transmission measurements, made with the Nigrosine in solid aerosol form, imply a $B_a$ of $3.7\text{m}^2/\text{g}$. The differences in these values represent a difference of less than 5%. The uncertainties in each of the values of $B_a$ are estimated to be approximately 10 to 15%, and so the values are considered to be in excellent agreement.

Measurements made with the DR technique for the nigrosine, however, showed a $B_a$ value of $0.9\text{m}^2/\text{g}$ at 633 nm, a value significantly lower than the DT and the aqueous solution value. The reasons for these differences are not known at this time. FR measurements were not made for the nigrosine.

The $B_a$ values at 633 nm were used to calculate $n_2$ values for the nigrosine. The aqueous transmission data results in a value of 0.25 for $n_2$; by comparison, a value of $n_2$ of 0.20 was inferred from in situ polar scattering data (A. Coletti, private communication).

Flo-Master Ink Measurements

Flow master ink, unlike the Mars ink consists entirely of material that is soluble in organic solvents. Like the nigrosine dye the ink forms a true solution. Flo-Master comparisons were made for DT, DR, and FR measurements. The solution measurements exhibited a
somewhat different wavelength dependence than the diffuse reflectance measurements, possibly due to differences between the Flo-Master ink as a solution or as solid aerosol particles. B_a values at 633 nm were determined to be 2.7 m^2/g for the DT data (with Nuclepore filters as substrates), 1.4 m^2/g for the DR data, and 0.6 m^2/g for the FR data. In this case as in the other cases investigated, use of the FR technique results in significantly lower B_a values than those measured by the other techniques.

Measurements with this ink were made to investigate the effect of varying filter media on diffuse transmission measurements. Results for the Nuclepore filter and the 0.45 μm Millipore filter are shown in Fig. 7 and Fig. 8. In addition, similar sets of measurements made with a 1.2 μm pore size Millipore filter and with fiberglass filters were quite similar to the data shown in Fig. 8. No enhancement of response was seen for any of these filters. These data, unlike previous data reported for soot aerosol, suggest little differences among the substrates. In particular, the differences in pore size for the two Millipore filters appeared to have no significant effect on the calculated values. Reasons for these inconsistencies between the data sets are not known at this time. The differences may, however, be related to differing penetration depths into the filters for the different materials.
Additional Comparisons

A further comparison of diffuse transmission and diffuse reflectance techniques for a sooty smoke emission from burning forest fuels is shown in Fig 2. In this figure, the solid line with circles and the solid line with triangles represent $B_a$ values determined for two components (one soluble in methylene chloride and the other non-soluble in methylene chloride) of the smoke emissions from flaming combustion; these measurements were made using a combination of DR and solution transmission techniques. The dashed line represents a weighted average of the two components. The $x$ represents a DT measurement of specific absorption in the flaming combustion aerosol. The DT data should lie on the weighted average line; the actual agreement is quite good. In this case, as in the others, the consistency in the different methodologies is excellent.

An additional comparison with volcanic ash also showed a consistency between the DT and the DR techniques. The comparison (for $n_2$ values) was made with an ash sample from El Chichon volcano. The $n_2$ values determined for these samples were $-0.0026$ for the diffuse transmission technique and $-0.0033$ for those measured by diffuse reflectance, numbers that are in agreement within about 25%. There are sample heterogeneities which add an uncertainty in these ash measurements due to sample variability. The uncertainty in these data is estimated to be approximately 20%, so that there is agreement within the uncertainties of the measurements.
SUMMARY AND CONCLUSIONS

Several sets of absorption measurements that include comparisons between different techniques for measuring submicron aerosol absorption have been made. These measurements have shown that there is a great deal of consistency among the different techniques when applied to well documented aerosols and that it is possible to provide some definite calibrations of the aerosol measurements in terms of aqueous transmission measurements. The measurements indicate (with the exception of the FR method) that there are no significant systematic errors in the data that are due to differences in techniques as applied in this study. Some specific conclusions follow:

1. It has been shown that the Mars ink is a useful analog of atmospheric soot for absorption measurements and that it can serve as a reproducible, stable aerosol for measurement.

2. The differences in measured $B_a$ values between aqueous and aerosol suspensions have been shown to be important in comparisons of different measurement techniques. Absorption values inferred in an aqueous suspension should be corrected to their values in air before use in aerosol studies.

3. It has been shown that, for our reference Mars ink carbon par-
articles, the diffuse transmission measurements and the diffuse reflectance measurements using the dilution technique give the $B_a$ value that is expected based on appropriately adjusted direct transmission measurements of the aqueous suspension. This provides a calibration of the diffuse transmission and diffuse reflectance methods in terms of more fundamental measurements.

4. It has been demonstrated that the filter reflectance method systematically underestimates the $B_a$ values of the highly absorbing samples. For the dilution method care must be taken to insure a uniform dilution for the sample.

5. Since the diffuse reflectance technique with dilution gives the expected results, the data indicate that the simple volume mixing rule used to calculate absorption for this method is adequate and that the use of Eq (5) is appropriate.
<table>
<thead>
<tr>
<th>Experimental Procedure</th>
<th>$B_a^2$ (m$^2$/g)</th>
<th>Best Fit Estimate</th>
<th>$n_2$ Eq (6)</th>
<th>$n_2$ Eq (10)</th>
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<td>Diffuse Reflectance</td>
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<td>Aqueous Suspension</td>
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Lindberg, J.D., and Snyder, D.G., Determination of the optical absorption coefficients of powdered materials whose particle size distribution and refractive indices are known, Appl Opt, 12, 573-578 (1973).


Fig. 1. Mie absorption efficiency plotted vs the Mie size parameter $x$. The solid line is the result of exact Mie calculations; the dashed line is the result of applying the approximation in Equation (10).

Fig. 2. Aerosol mass distribution determined for the Flo-Master ink. The upper curve is a mass distribution determined from scanning electron micrographs; the lower curve from a single particle optical counter. Although the upper curve extends only to 0.5 μm radius, an inspection of the micrographs showed no significant mass in the larger size ranges.
Fig. 3. Measurements of the turbidity of a Mars ink aqueous suspension plotted against mass concentration in a 1 cm path length optical cell.

Fig. 4. Measurements of the turbidity of a Mars ink aerosol plotted against mass concentration on a 14 cm² Nuclepore filter.
Fig. 5. A comparison of $B$ values determined for the Mars ink by different measurement techniques. The agreement among the techniques is excellent, both in the absolute values and in the wavelength dependence.

Fig. 6. Measurements of the turbidity of a Nigrosine solution plotted against the Nigrosine concentration.
Fig. 7. Measurements of the turbidity of a Nigrosine aerosol collected on a 14 cm² Nuclepore filter vs the mass on the filter.

Fig. 8. Measurements of the turbidity of a Flo-Master ink aerosol collected on a 14 cm² Nuclepore filter vs the mass on the filter.
Figure 9. Measurements of the turbidity of a Flo-Master ink aerosol collected on a 14 cm² Millipore filter vs the mass of aerosol on the filter.

Figure 10. Measurements of the absorption properties of smoke emissions from flaming combustion. The solid line with circles (-o--o-) represents the wavelength dependent $B_m$ measured for the non-soluble component determined by diffuse reflectance. The lower solid line with triangles(-Δ--Δ-) represents the $B_m$ measured for the soluble component by solution transmission. The dashed line represents a weighted average of the two components. The $x$ represents a measurement made by the diffuse transmission technique for a total filter sample. The upper solid line is the $B_m$ expected for a pure graphitic carbon aerosol.