

**On the Wavelength Dependence of
the Absorption of Black Carbon Particles:
Predictions and Results from the TARFOX Experiment
and Implications for the Aerosol Single Scattering Albedo**

By

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Abstract

We present measurements of the wavelength dependence of the aerosol absorption coefficient taken during the TARFOX experiment over the Northern Atlantic. The data show an approximate λ^{-1} variation between 0.40 μm and 1.0 μm . We explore the theoretical basis of the wavelength variation of the absorption of solar radiation by elemental carbon (or black carbon, BC). For a wavelength independent refractive index the small particle absorption limit simplifies to a λ^{-1} variation in relatively good agreement with the data. This result implies that the refractive indices of BC were relatively constant in this wavelength region, in agreement with much of the data on refractive indices of BC. However, the result does not indicate the magnitude of the refractive indices.

We discuss the implications of the wavelength dependence of BC absorption for the spectral behavior of the aerosol single scattering albedo. We show that the single scattering albedo for a mixture of BC and non-absorbing material decreases with wavelength in the solar spectrum (i.e., the percentage amount of absorption increases). This decrease in the single scattering albedo with wavelength for black carbon mixtures is different from the increase in single scattering albedo for most mineral aerosols (dusts). This indicates that if generally true the spectral variation of the single scattering albedo can be used to distinguish aerosol types. It also highlights the importance of measurements of the spectral variation of the aerosol absorption coefficient and single scattering albedo.

1. Introduction

Absorption of solar radiation by black carbon (BC) particles is important in understanding the effects of atmospheric particles on climate. Particles with no absorption have a negative (cooling) forcing while particles with substantial absorption can have a positive (warming) forcing. As Heintzenberg et al. (1997) noted, “Even though BC constitutes only a few percent of the aerosol mass, it can have a significant positive forcing.” Simple calculations show that a small amount of BC can change a negative aerosol forcing to a positive aerosol forcing. Jacobson recently found that the magnitude of the direct radiative forcing from black carbon itself exceeds that due to CH₄ and may be the second most important component of global warming after CO₂ in terms of direct forcing (Jacobson, 2001a). In addition to the direct climate effect, the INDOEX experiment recently showed that BC particles were apparently responsible for reduced cloudiness over the Indian Ocean (Ackerman et al., 2000).

Black carbon particles have received increased interest recently since Hansen et al (2000) indicate that it is perhaps more cost effective to control black carbon particles than CO₂ at the present time. However, before embarking on a large scale control strategy for black carbon particles, it is important to fully understand their radiative properties and effects.

Black carbon particles enter the atmosphere as a result of incomplete combustion processes, whether from motor vehicles, industrial flames, or biomass burning. If the flame temperature is too low or there is a limited supply of oxygen, not all of the fuel is converted to carbon oxides and carbon soot results. As combustion is ubiquitous, carbon soot is found virtually everywhere. (e.g., off the eastern coast of US, Novakov et al (1997); off the western coast of Europe, Putaud et al. (2000), Novakov et al. (2000); off the Indian Peninsula, Ramanathan et al. (1996); in China, Chameides et al. (1999); in Brazil, Kaufman et al. (1999); over the Pacific Ocean, Kaneyasu and Murayama (2000) and in remote areas, Bodhaine (1995)). Global simulations of carbonaceous aerosols show a worldwide distribution of BC with maxima over the heavily industrialized areas and biomass burning regions (Liousse et al., 1996).

In this paper we present measurements of the wavelength dependence of the aerosol absorption coefficient due to BC obtained during the TARFOX experiment. We then discuss the effects of particle size and refractive indices on the absorption coefficient of BC and, in particular, the wavelength dependence of the absorption coefficient. The data show that the

absorption seen in the TARFOX experiment has an approximate λ^{-1} dependence that is in good agreement with the small particle limit for BC particles for a constant refractive index.

We also briefly discuss the implications of the results in terms of the single scattering albedo of atmospheric aerosols. We show that for situations where the absorption is due to BC, the single scattering albedo of the aerosol mixture should decrease with wavelength. This is in contrast to most mineral aerosols (dusts) where the single scattering albedo usually increases with wavelength (Sokolik and Toon, 1997; 1999).

2. Measurements of the aerosol absorption coefficient during the TARFOX experiment.

The TARFOX field program was carried out off the Eastern Coast of the United States in the summer of 1996 (Russell et al., 1999a). Samples of the aerosol were obtained aboard the UK Met Office Hercules Aircraft (C130) on three particular days; July 21, July 25 and July 27, 1996. The filters were exposed during the level passes when the solar radiation values were measured (Hignett et al., 1999). The samples were taken back to the UK Met Office laboratory and the absorption coefficients at wavelengths between 0.45 μm and 1.0 μm were measured using the integrating sandwich method (Clarke, 1982; Foot and Kilsby, 1989).

2.1 Integrating Sandwich method

The integrating sandwich technique was developed to minimize the influence of the scattering properties of the measured material on the absorption determination (Clarke, 1982). The material is placed between two high-reflectance diffusing wafers and the resultant attenuation of light is measured. The method is discussed in detail elsewhere (Clarke, 1982; Foot and Kilsby, 1989; Horvath, 1993 and Heintzenberg et al., 1997). Clarke (1982) estimated the error to be +/- 25%.

The major uncertainties in the method arise from the additional scattering in the layer from other particles (Horvath, 1997) and knowing how many passes the light makes through the media. This causes additional apparent absorption and leads to an overestimation of the absorption coefficient. However, in this study we are concerned with the wavelength dependence of the absorption coefficient and not the absolute magnitude of the absorption coefficient. We have included the standard deviation of the individual measurements as a

measure of the intrinsic variability similar to Hignett et al. (1999) who showed some of this data in their Figure 4.

2.2 Absorption Coefficient Results

The measured absorption coefficients are shown in Figure 1. Only the data when the airplane was in the aerosol layers are plotted. The relative standard deviations are larger at longer wavelengths and for smaller absorption coefficients. To isolate the wavelength dependence, the data are normalized by the absorption coefficient at 0.55 μm and shown in Figure 2. The results show clearly the decrease in absorption coefficient with wavelength. As seen, the wavelength variation on the three days is roughly the same. The absorption decreases quite quickly with wavelength between 0.4 and 0.6 μm and then levels off at the longer wavelengths. There is a relatively large spread in the value at 1.0 μm .

The aerosol conditions on two of the TARFOX days were quite similar (July 25 and 27th), but the conditions on July 21st were very different (low optical depth on the 21st but very hazy on July 25 and 27th). The fact that the wavelength dependence is very similar on all three days is notable and possibly indicates that the absorbing material has a common origin.

The aerosol absorption observed in TARFOX was attributed to BC (Novakov et al., 1997; Hegg et al., 1999) since the correlation of the absorption coefficient with measured carbonaceous aerosol amount was quite high ($r^2 = 0.69$) and the observed absorption coefficient indicated roughly 10% of the carbonaceous aerosol was BC. As we do not have detailed chemical information on the aerosol encountered in the TARFOX experiment, it is possible that there were other absorbing hydrocarbons present. Jacobson has pointed out that other materials such as benzene absorb in the UV in a similar manner as BC particles (Jacobson, 2001b). However, for simplicity, we will refer to the absorption measured in the samples as absorption due to BC particles.

Since the samples were collected on filters, the absorption measurements are for the dry aerosol only. We do not consider here the effects of water or other coatings on the particles (Ackerman and Toon, 1980; Fuller et al., 1999; Redemann et al., 2000). While important for the absolute magnitude of the absorption, these effects probably have less influence on the wavelength dependence of the absorption coefficient. Jacobson (2000) shows some sample calculations for externally-mixed, internally-mixed and shell-core configurations of BC. While

the magnitudes of the calculated absorption coefficients differ, the wavelength dependence is very similar for all three cases for the spectral region we are concerned with here (0.45 – 1.0 μm).

3. Calculations of the Absorption Coefficient of BC

The optical properties of BC have been studied in the context of industrial flames (Dalzell and Sarofim (1969), Lee and Tien, (1981), Chang and Charalampopoulos, (1991), Stagg and Charalampopoulos (1993)), paint pigments (Medalia and Richards (1972)), urban atmospheres (Horvath, 1993, Horvath et al., 1997), the nuclear winter debate (Turco et al., 1991; Golitsyn, et al., 1988; Penner, 1986) and more recently, biomass burning (Kaufman et al., 1999, Reid et al., 1999).

Fuller et al. (1999) have recently explored in some detail the effects of various sizes, shapes and configurations on the extinction of light by BC particles. They considered the effects of material composition and morphology of soot and the electrodynamics of spherules agglomerated into loose (ramiform) aggregates. They found a wide variation in the mass absorption coefficient at 0.55 μm depending upon the variations in optical constants. Their results suggest the importance of determining the physical state of the soot particles and the immediate environment when ascribing characteristic values for the absorption and scattering efficiencies.

However, Fuller et al. (1999) did not directly address the wavelength dependence of the absorption by BC particles and our results suggest that a less detailed analysis may be sufficient for explanation of the wavelength dependence of the absorption. Therefore, we discuss here only the small particle limit and the effect of the refractive indices on the absorption coefficient.

3.1 Size effects

In general, the radiative properties of atmospheric aerosols are a strong function of size. Electron microscope analyses show that most of the total BC mass is composed of fractal agglomerates made up of primary spherules with a diameter of 0.03 – 0.05 μm (Seinfeld and Pandis (1998) Fig. 13.1; Martins et al., (1998)). For very small spherical absorbing particles (roughly $r < 0.01 \mu\text{m}$ for $\lambda = 0.55 \mu\text{m}$) the absorption per mass (specific absorption or mass absorption coefficient) becomes independent of size (van de Hulst, 1957; Bergstrom, 1973;

Bohren and Huffman, 1983; Horvath, 1993; Colbeck et al., 1989; Martins et al., 1998). This size-independent specific absorption coefficient relationship corresponds to the so-called Rayleigh absorption limit of

$$\kappa = (\text{constant}/\rho\lambda)\text{Im}[(m-1/m-2)] = (\text{constant}/\rho\lambda) nk/((n^2-k^2+2)^2+(2nk)^2) \quad (1)$$

where κ is the specific or mass absorption coefficient, m is the complex index of refraction ($m = n - ik$) and ρ is the density of the particle. An important result of Equation 1 is that for small particles and a constant refractive index the wavelength dependence of the mass absorption coefficient is simply λ^{-1} .

Equation 1 is only strictly applicable to absorption by a spherical particle with a uniform index of refraction. Berry and Percival (1986) developed a fractal theory to describe the absorption by the BC fractal agglomerates. This theory also predicts that the specific absorption of these agglomerates should not be a strong function of size. (However, it is a strong function of the optical constants and the physical state of the particle, Fuller et al 1999.) Experimental results are in good agreement with the fractal model predictions (Colbeck, 1989; Petzold et al., 1997 figure 5)

Data from Horvath (1993) do show a decrease in the specific absorption with size but Petzold et al. (1997) argue that these larger black carbon particles that are not fractal clusters (and hence have less specific absorption) are a small amount of the total BC mass.

For very large spherical particles the absorption (again, for constant refractive index) becomes wavelength independent (Bergstrom, 1973). Thus we can infer that the wavelength dependence of the absorption coefficient of a BC particle with a constant refractive index should be about λ^{-1} to $\lambda^{-1.3}$ or less. (The Angstrom exponent for absorption by a BC particle with effective radius of ~ 0.02 μm is 1.3 at 0.55 μm .) This result is independent of what the absolute value of the refractive index is (or the magnitude of the specific absorption).

3.2 Refractive index of BC

It is well established that the refractive indices of BC particles vary greatly depending upon the details of the combustion process (fuel type, flame temperature, etc). There is a wide variation in the reported value of refractive indices of BC (Twitty and Weinman, (1972) Figure

1; Colbeck et al., 1989 Table 1; Fuller et al., 1999). Bohren (1982) noted that there is about a factor of 5 variation in the imaginary part of the index of refraction for what is called “soot.”

The wide variation in the measured refractive indices most likely comes from the amount of other hydrocarbons contained in the particles. Batten (1985) notes that the imaginary part of the refractive index is inversely correlated with the H/C ratio. In other words, the closer the particle is to pure carbon (i.e. graphite) the larger the refractive indices. Also, as mentioned above, other hydrocarbon compounds may absorb in a similar manner to BC further complicating the measurement of the refractive indices.

Undoubtedly, there is a large variation in the absorption coefficient of the carbon particles that exist in the atmosphere depending upon their sources and the changes that occur while in the atmosphere (coatings, chemical reactions). For example, Fuller et al. (1999) classified BC into 5 different categories with different refractive indices and densities for each group.

To explore the effect of the refractive index on the wavelength dependence of the absorption, we show two different reported values for the spectral variation of the refractive indices of flame soots (Lee and Tien, 1981 and Chang and Charalampopoulos, 1990) in Figure 3. Both studies represented the molecular structure of the soot as graphite (i.e. 3 bound electrons and one free electron). Then, they computed values for the damped harmonic oscillator, etc. in the Kramers - Kroenig formula (Bohren and Huffman, 1983). They fit their transmission data and came up with the index of refraction as a function of wavelength for the particular type of flame soot being studied.

The wavelength dependence of the refractive indices of Lee and Tien is larger than for the Chang and Charalampopoulos values. The imaginary part of the Lee and Tien refractive index varies from 1.0 at 0.3 μm to a minimum of 0.38 at 0.5 μm and then increases to 0.9 at 1.0 μm . By contrast, the Chang and Charalampopoulos refractive indices have relatively little wavelength variation similar to other estimates of the refractive indices of flame soot (Dalzell and Sarofim, 1969; Bergstrom, 1972). It should be emphasized that different flame soots (acetylene, propane, diesel fuel, etc) have different values for the refractive indices. These two choices of the refractive indices illustrate the range in the wavelength dependence in the region of 0.3 to 1.0 μm from relatively constant to varying by more than a factor of 2.

The wavelength variation in the Lee and Tien refractive indices results from their expression for the temperature dependence of the electron damping coefficients. Lee and Tien show this temperature dependence in their Figure 5 where the variation of the imaginary index with wavelength increases significantly as the temperature decreases. Stagg and Charalampopoulos (1993) indicated that there was no significant temperature variation in their determined refractive indices of flame soot. They question the use of the temperature dependence of the electron damping coefficients used by Lee and Tien (1981).

In the UV the increase in refractive indices is termed a "glassy carbon" resonance by Chang and Charalampopoulos (see also Bruce et al., 1991). This absorption could be important for photochemical reactions.

At the longer wavelengths (past 1.0 μm), the refractive indices of BC also increase with wavelength. This is due to properties of the free electron and becomes important for the thermal emission properties of flame soot (Lee and Tien, 1981). As mentioned above, the absolute magnitude of refractive indices varies greatly depending upon the type of fuel and specific combustion processes.

4. Comparison of predicted and measured absorption and discussion

4.1 Comparison to the TARFOX measurements

Using the values of the wavelength dependence of the refractive indices discussed above we computed the value of specific absorption from Eq.(1) and plotted the relative values in Figure 4. Also plotted is a simple λ^{-1} curve that represents a constant refractive index.

To describe the goodness of fit of the computed values to the data we use two measures: reduced chi-squared, χ_v^2 , and the root-mean-square difference between computed and measured values. χ_v^2 (e.g. Bevington and Robinson, 1992) is proportional to the weighted sum of squares of the difference between each data point and the corresponding computed value; the weight for each difference is the inverse square of its measurement error bar. As noted by Bevington and Robinson (1992), if the calculated curve is a good approximation to the data, χ_v^2 should be approximately unity or less. Increasingly larger values indicate increasingly poorer fit. χ_v^2 is 0.79 for the Chang and Charalampopoulos refractive indices, 1.97 for the constant refractive

index (λ^{-1}) and 38.23 for the Lee and Tien refractive indices. Corresponding rms differences between computed and measured values are 0,085, 0.12, and 0.49.

As shown in Figure 4 the λ^{-1} curve (the small particle limit with constant index of refraction) fits the data within nearly all the uncertainty bars ($\chi_v^2=1.97$). The wavelength dependence of the absorption coefficient using the Chang and Charalampopoulos refractive indices is very similar to that obtained using a constant index of refraction and in fact fits the data somewhat better ($\chi_v^2=0.79$). Use of the Lee and Tien refractive indices, however, overestimates the relative absorption at wavelengths longer than 0.6 μm and gives a very poor fit overall ($\chi_v^2=38$). However, the Lee and Tien refractive indices do capture the flattening out of the absorption at longer wavelengths.

Our results indicate that the simple constant index of refraction - small particle limit is a relatively good representation of the wavelength dependence of the aerosol absorption by BC observed in the TARFOX field campaign. This implies that the refractive indices of BC were relatively constant in agreement with much of the data on BC (such as Chang and Charalampopoulos, 1990)

4.2 Comparison to other Measurements of BC Specific Absorption

There is only a limited amount of data on the spectral absorption coefficient of BC particles (as compared to the amount of data on the specific absorption in the mid-visible, 0.50 – 0.55 μm). Bruce et al. (1991) compared the available spectral data for the absorption and scattering properties of diesel soots. They found that there was an approximate λ^{-1} dependence at selected laser wavelengths from 0.488 μm to 0.857 μm .

Horvath et al. (1997) found a wavelength dependence of $\lambda^{-0.92}$ for the aerosol absorption from their studies in Santiago, Chile. The absorption was most likely due to BC particles since the measurements were made in an urban area.

Moosmuller et al. (1998) measured the absorption coefficient of atmospheric aerosols near Denver, CO at 0.532 μm and 0.685 μm with a photoacoustic instrument. They compared their results with other measurements that indicated a wavelength dependence of the absorption of $\lambda^{-2.7}$. However there were only a few measurements included and from different sources. Also shown in Fig 4 is the $\lambda^{-2.7}$ curve. It clearly underestimates the TARFOX absorption for wavelengths greater than about 0.6 μm .

Dubovik et al. (1998) inferred the aerosol absorption optical depth of the atmosphere from sky radiance data for areas that have significant biomass burning. Their results show that for one year the wavelength dependence was λ^{-1} but for two other years the wavelength dependence was greater than λ^{-2} . Dubovik et al. (1998) argue that the cause of the increase in wavelength dependence of the absorption optical depth is most likely due to the wavelength dependence of the imaginary refractive indices (assuming the small particle limit is valid). However, as discussed above, the data from flame soots do not indicate that the imaginary index decreases enough from 0.3 – 1.0 μm to account for a λ^{-2} relationship. It is more likely that minerals (dusts) in the atmospheric aerosol caused the more rapid decrease in absorption with wavelength (see below). Dubovik et al. (1998) noted that iron oxide has refractive indices that would cause such an increase in the wavelength dependence. Another possibility is that the biomass burning aerosol is sufficiently complicated (see Martins et al., 1999) that at times the λ^{-1} relationship is not applicable.

While combustion processes vary widely from place to place (eg., diesel exhaust to biomass burning), it would appear that the small BC particles may have a common structure and perhaps a common wavelength dependence of the absorption. Such a result would be very useful for the estimate of the climatic impact of BC particles.

4.3 Implications for the single scattering albedo of the atmospheric aerosol

The single scattering albedo is a critical parameter for determining the effect of aerosols on the global temperature. It is defined as the ratio of the scattering to the extinction, i.e.

$$\omega = \sigma/\beta$$

where σ is the scattering coefficient and β is the extinction coefficient. Or expressed in terms of the absorption coefficient

$$\omega = 1-(\kappa/\beta)$$

If there is a mixture of small BC (or BC-like material) particles and other non-absorbing particles (such as sulfates), then the non-absorbing particles will dominate the scattering and the BC the absorption. If the extinction also follows a "power law" then

$$\omega = 1 - (\kappa_0/\beta_0)(\lambda/\lambda_0)^{-1+\alpha} \quad \text{where } \alpha \text{ is the Angstrom exponent} \quad (2)$$

For atmospheric aerosols that do not contain large particles (i.e. dust) α usually is greater than 1, so the single scattering albedo of the mixture should decrease with wavelength. This is due to the fact that the scattering coefficient usually decreases with wavelength faster than the absorption coefficient (Bergstrom 1973). As the scattering coefficient goes to zero, the extinction coefficient equals the absorption coefficient and the single scattering albedo is zero. This result is also discussed by Dubovik et al. (1998).

However, for BC particle clusters, the single scattering albedo does not drop to zero at visible wavelengths. Bruce et al (1991) show that the single scattering albedo for BC clusters is about 0.3 for 0.554 μm . This apparently is due to the fact that the scattering coefficient of BC clusters does not follow the small particle limit (which is Rayleigh scattering). Berry and Percival's fractal theory indicates that the scattering cross-section for the cluster is larger than that for the sum of the spherules (unlike the absorption coefficient). However, Bruce et al (1991) do show that the single scattering albedo for carbon particles does get very small at 100 μm .

Estimates of the TARFOX aerosol single scattering albedo based on model calculations do show a decrease with wavelength (Hignett et al., 1999; Russell et al., 1999b). Estimates of the effects of water growth on the particles do not change the decrease with wavelength (of course, the single scattering albedo increases with relative humidity due to the water taken up by the particles – See Hignett et al. (1999) Figure 3).

Recently, the SCAR-B program measured the optical properties of the aerosol from biomass burning in Brazil (Kaufman et al., 1999). The biomass burning BC particles have a very complicated structure and are difficult to describe simply (see Martins et al., 1998 Figure 5). However, Reid and Hobbs (1998) found that the single scattering albedo decreased with wavelength as did Eck et al. (1998).

In contrast to BC, large particles (dust or sea salt) usually have an Angstrom coefficient less than one (or relatively constant optical depth in the solar spectrum, Sokolik and Toon

(1997). In addition, the small particle absorption limit of Eq 1 is not valid for these large particles. However, it is useful to compare the wavelength dependence of the single scattering albedo of mineral aerosols and BC. Sokolik and Toon (1997) show single scattering albedos for various dust measurements and Sokolik and Toon (1999) present the refractive indices and radiative properties for a number of mineralogical components. Their results indicate that the single scattering albedo for dust increases with wavelength in the range of 0.3 to 1.0 μm . This is due to significant absorption in the UV and visible wavelengths less than $\sim 0.6 \mu\text{m}$ due to electronic bands and less absorption at longer solar wavelengths. Because the extinction coefficient is relatively constant with wavelength, the change in single scattering albedo is determined simply by the absorption coefficient spectral behavior.

If the difference in wavelength behavior between BC and dust is generally true, it means that the wavelength dependence of the single scattering albedo could be used to differentiate between dust absorption and BC absorption.

5. Conclusion

We have analyzed the wavelength dependence of the absorption coefficient measurements of aerosol collected during the TARFOX field campaign. The results show a decrease in the absorption of λ^{-1} that is consistent with the absorption of small particle BC. This result implies that the refractive indices of BC in this spectral region were relatively constant in agreement with much of the data on the refractive indices of BC.

We show that for atmospheric aerosol mixtures where the absorption is due to BC and the extinction wavelength dependence is less than λ^{-1} , the single scattering albedo should decrease with wavelength in the region of 0.3 to 1.0 μm . This is in agreement with the limited data available. It contrasts with the increase in the single scattering albedo for mineralogical dusts and indicates the need for more careful measurements of the wavelength dependence of the aerosol absorption and the single scattering albedo.

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Figure Captions

Figure 1: Aerosol absorption coefficients as a function of wavelength for three TARFOX days, July 21 – a, July 25 – b, and July 27 – c.

Figure 2: Aerosol absorption coefficients as a function of wavelength normalized by the value at 0.5 μm – a, July 25 – b, and July 27 – c.

Figure 3: Refractive indices for flame soots from Lee and Tien (1981) and Chang and Charalampopoulos (1990)

Figure 4: Normalized aerosol absorption coefficients as a function of wavelength. Also shown is a λ^{-1} curve and calculations using equation (1) with the Lee and Tien and Chang and Charalampopoulos refractive indices. The chi-squared value for the Chang and Charalampopoulos refractive indices is 0.79 (rms error 0.085), 1.97 for the constant refractive index (rms error 0.12) and 38.23 for the Lee and Tien refractive indices (rms error 0.49)

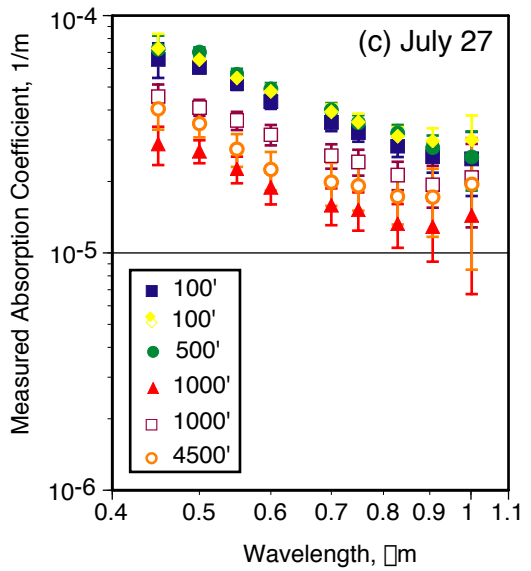
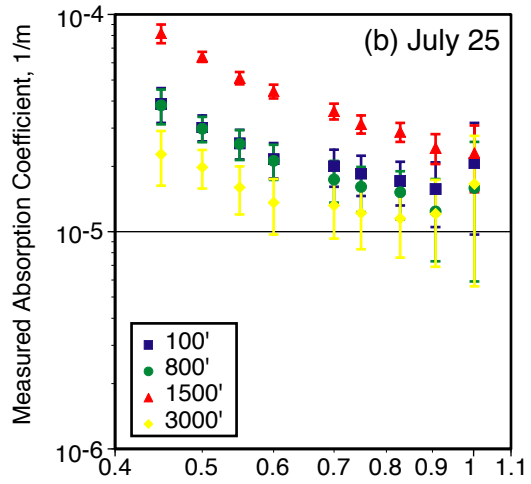
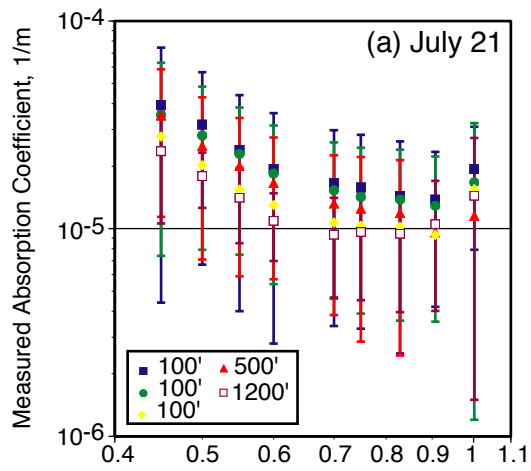


Figure 1

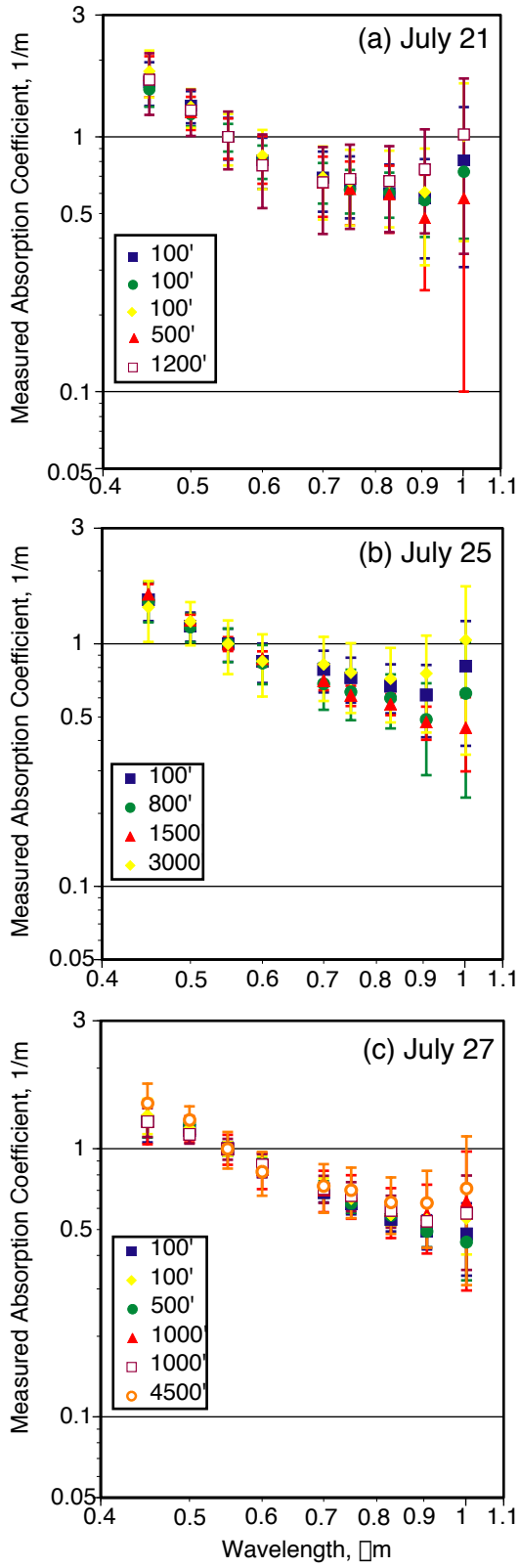


Figure 2

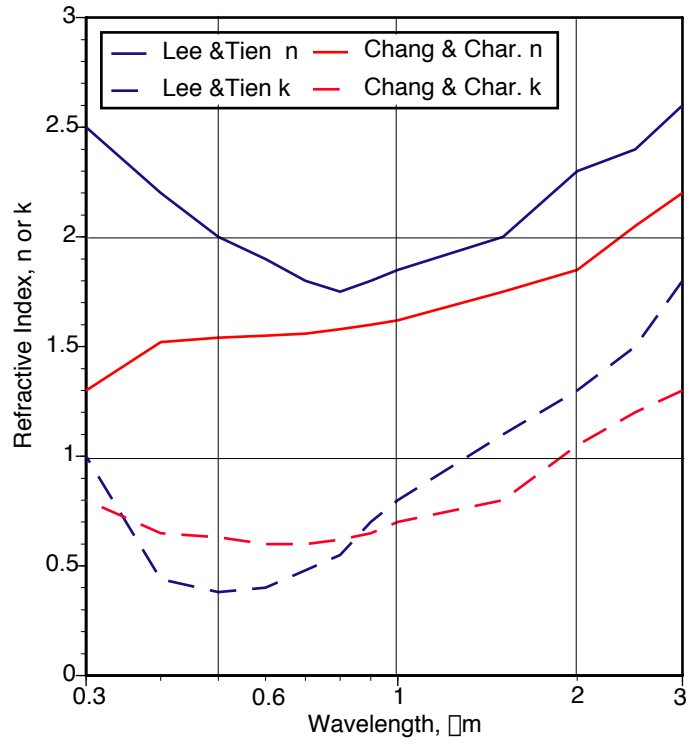


Figure 3

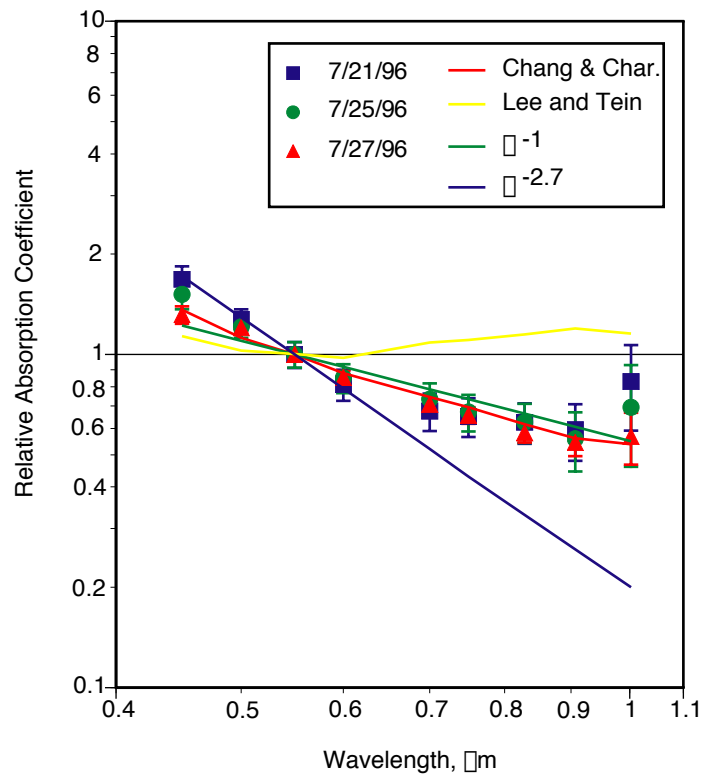


Figure 4