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H. Rosen and T. Novakov

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OPTICAL TRANSMISSION THROUGH AEROSOL DEPOSITS ON DIFFUSELY REFLECTIVE FILTERS: A METHOD FOR MEASURING THE ABSORBING COMPONENT OF AEROSOL PARTICLES*

H. Rosen and T. Novakov

Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

Recent studies of aerosol particles in urban, rural, and remote environments show the presence of substantial concentrations of combustion-generated graphitic carbon particles.[1-3] Associated with these graphitic particles are large optical absorption coefficients, which could significantly affect radiation transfer through the atmosphere.[4] Although a few in-situ measurements of this component have been reported,[5-7] the predominant measurement technique involves collecting particles on a filter medium and determining the optical transmission of the deposit.[1,8] A laser transmission technique developed at Lawrence Berkeley Laboratory has been used extensively to determine the absorption coefficient and the graphitic carbon content of aerosols in the United States,[9] some areas of Europe, and the Arctic region.[2] The laser transmission apparatus compares the transmission

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of a 632.8-nm He-Ne laser beam through a loaded filter relative to the transmission through a blank filter, as shown in Fig. 1. After multiple scattering through the filter medium, the forward scattered light is collected by low f-number optics and detected by a photomultiplier. The filters used in our method are almost perfect diffuse reflectors, and only a small fraction of the incident beam is transmitted through them.

This technique appears to be a direct measure of the absorbing component of aerosol particles and is insensitive to its scattering properties. This has been shown directly in a photoacoustic study[10] and indirectly by noting that most of the aerosol mass responsible for scattering can be removed by solvent extraction or heat treatment procedures without appreciably affecting the optical transmission measurement.[11] Furthermore, it has been shown that adding nonabsorbing particles to the filter substrate does not give an appreciable optical attenuation.[12] From a theoretical point of view, these results are not obvious, since it is unclear why the backscattered radiation from nonabsorbing particles should not make a significant contribution to the optical attenuation measurement. This is especially true where the absorbing component represents only a very small fraction of the aerosol mass. In this paper, we will present a simple theoretical model which accounts for all these observations and points out the critical role of the filter substrate as an

almost perfect diffuse reflector in the technique. Similar considerations may also apply to the opal glass method developed by Lin et al.[8]

For this model calculation, we will assume that the particles and the filter media can be treated independently and consider the geometry shown in Fig. 2. A similar treatment, where the light beam is first incident on the particles, gives identical results. After the light beam passes through the filter medium, it is incident on the particles with an intensity, I_0. The particles forward scatter a fraction of the incident light, backward scatter a fraction, and absorb a fraction. These components in the low loading limit are respectively given by $n_s\sigma_F I_0$, $n_s\sigma_B I_0$, and $n_A\sigma_A I_0$, where n_s is the number of scattering aerosol particles per unit area, n_A, is the number of absorbing aerosols per unit area, σ_F is the forward scattering cross section, σ_B is the backward scattering cross section, and σ_A is the absorption cross section.

Since the optical attenuation technique only measures the forward scattering light, it would seem as if the backscattered light would be lost to the system and would contribute to the attenuation. However, the filter, in our method, is almost a perfect reflector. Under these circumstances, the backscattered light will be reflected in the forward direction and will again be incident on the particles. This process will continue until almost all the backscattered radiation is collected by the optics and therefore does not contribute to the optical attenuation. This result can be put in

mathematical form, where I is the light detected by the collection optics and R_F is the reflectivity of the substrate.

$$I = I_0(1 - n_s\sigma_b - n_A\sigma_A) + I_0(1 - n_s\sigma_B - n_A\sigma_A)n_s\sigma_B R_F \qquad (1)$$
$$+ I_0(1 - n_s\sigma_B - n_A\sigma_A)n_s^2\sigma_B^2 R_F^2 + \ldots I_0(1 - n_s\sigma_B - n_A\sigma_A)n_s^n\sigma_B^n R_F^n$$
$$= I_0 \frac{(1 - n_s\sigma_B - n_A\sigma_A)}{(1 - n_s\sigma_B R_F)} .$$

Consider several limits. Where $R_F \simeq 0$, which normally would be considered an ideal substrate, Eq. (1) reduces to

$$I = I_0(1 - n_s\sigma_B - n_A\sigma_A) .$$

Under these conditions, the backscattered radiation will contribute significantly to the optical attenuation and make the technique unsuitable for exclusively measuring the absorbing properties of the aerosol. In our method, however, $R_F \simeq 1$ and Eq. (1) becomes

$$I = I_0(1 - \frac{n_A\sigma_A}{1 - n_s\sigma_B}) .$$

From this expression it is clear that a nonabsorbing aerosol will not contribute to the optical attenuation; this is consistent with our experimental results. The magnitude of the optical attenuation is somewhat dependent on the scattering properties of the aerosol; however, in the low loading limit, this effect is small. For example, if the substrate has 50% coverage, the scattering cross section of the particles, σ_s, is twice the particle area, and σ_B is 20% of σ_s. Then

$$1 - n_s \sigma_B \simeq .8 \ .$$

This implies that even for these large scattering cross sections and rather high loading, the error in the absorption measurement due to scattering of the aerosol is only about 20%. This treatment should only be viewed as giving physical insight into why transmission measurements through aerosol particles collected on highly reflecting substrates selectively measures the absorbing component of the aerosol particles. It is clearly approximate since it assumes that the optical properties of the particles are not affected by the filter substrate and neglects penetration of the particles into the substrate, which can lead to enhanced absorption due to multiple scattering within the filter media. These effects could be substantial and require further investigation.

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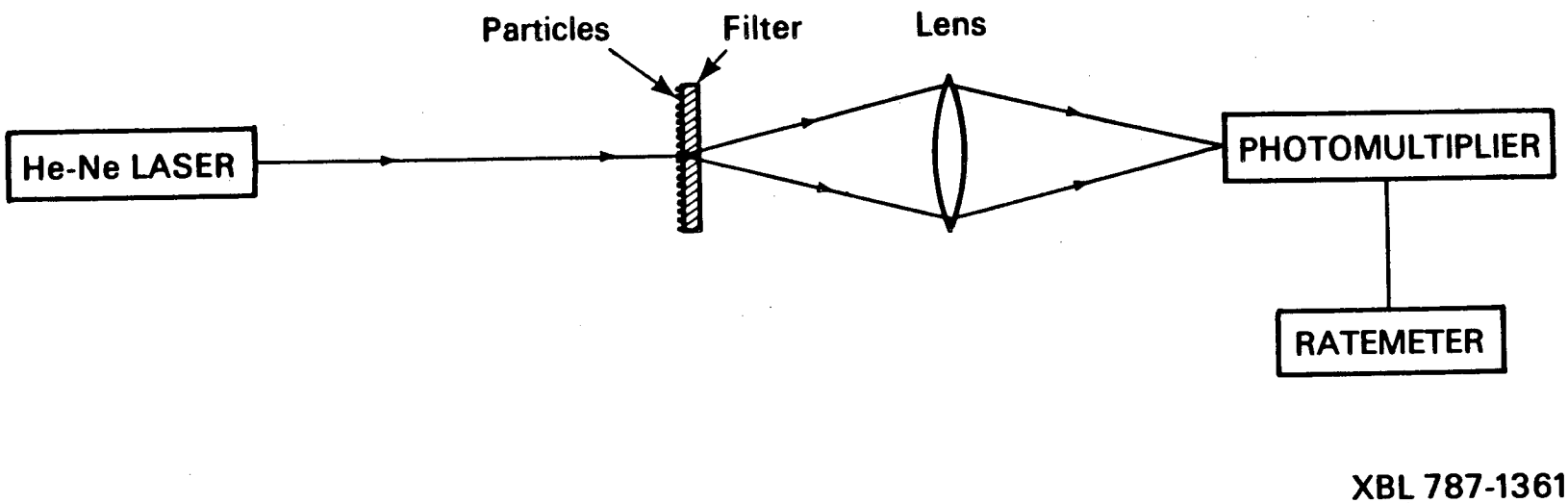
Figure Captions

Figure 1. Schematic of the Lawrence Berkeley Laboratory laser transmission apparatus.

Figure 2. Schematic of the experimental arrangement used for the model calculation.



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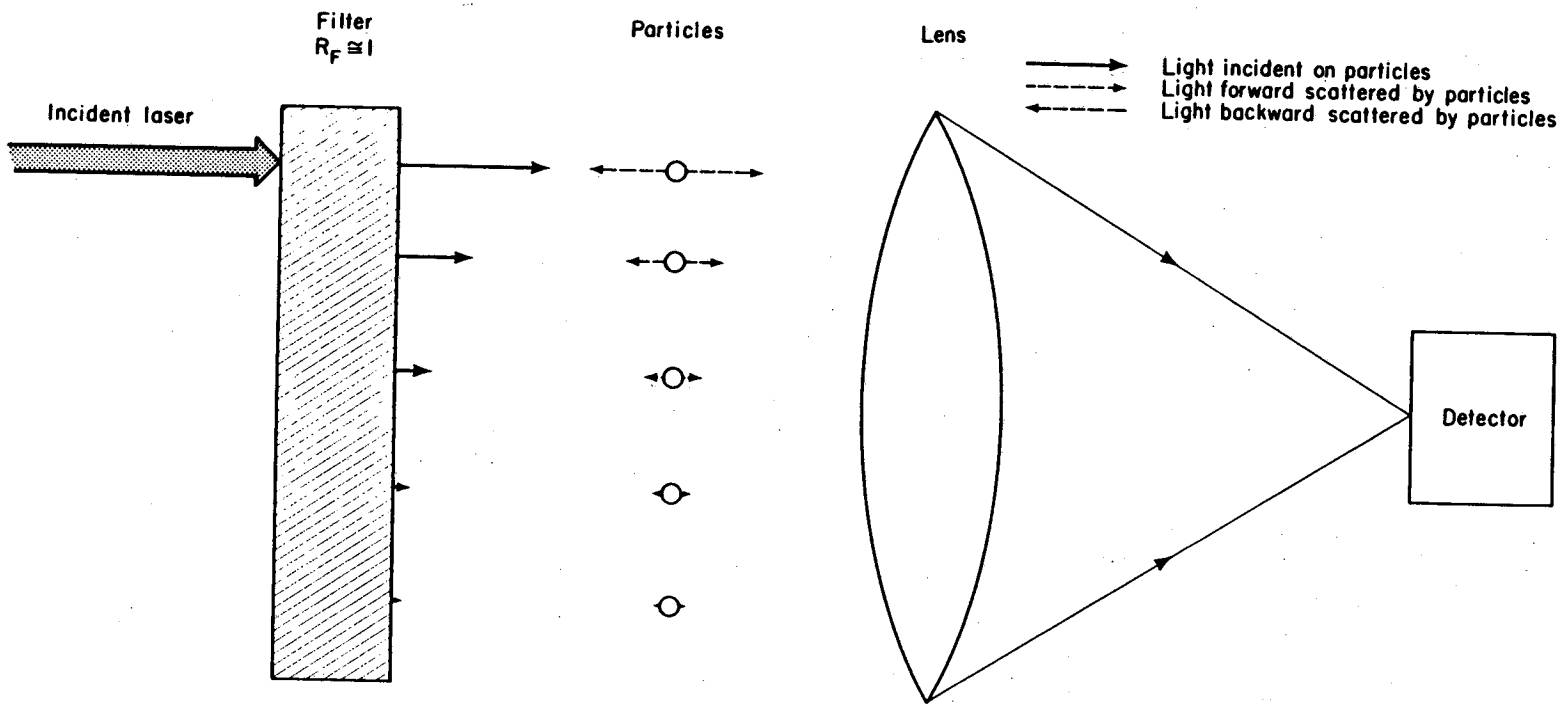

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Figure 2. Schematic of the experimental arrangement used for the model calculation.

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