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LAWRENCE BERKELEY LABORATORY LASER TRANSMISSION METHOD

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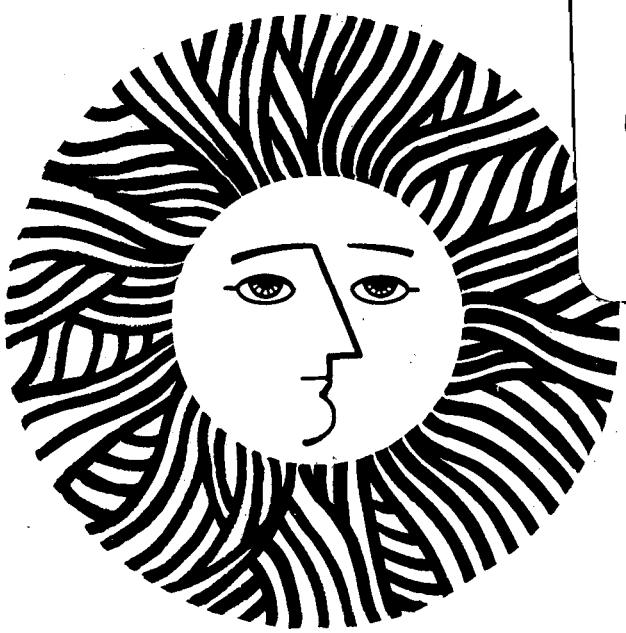

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LAWRENCE BERKELEY LABORATORY LASER TRANSMISSION METHOD*

H. Rosen and T. Novakov

Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

Abstract

The Lawrence Berkeley Laboratory laser transmission method has been used to determine the absorption coefficients of aerosol particles generated at the First International Workshop on Light Absorption. Analysis of the results confirms that the optical attenuation measurement is insensitive to the scattering properties of the aerosol. A simple model calculation is presented which explains these observations and points out the critical role of the filter substrate as an almost perfect diffuse reflector in the technique.

*This work was supported by the Assistant Secretary for the Environment, Office of Health and Environmental Research, Pollutant Characterization and Safety Research Division of the U.S. Department of Energy under Contract No. W-7405-ENG-48, and by the National Science Foundation, Grant No. ATM 80-13707.

Introduction

An optical attenuation technique has been developed at Lawrence Berkeley Laboratory (LBL) to determine the absorption coefficient of aerosol particles.[1] This technique has been applied on a routine basis to the analysis of filter samples collected in many urban[2] and rural areas in the United States, several sites in Europe, and in the Arctic region.[3]

The LBL technique compares the transmission of a 633-nm He-Ne laser beam through a loaded filter (Millipore, quartz, and certain other filter media) relative to that of a blank filter (Fig. 1). The loaded filters are placed in the beam with the loaded side towards the laser; after multiple scattering through the filter substrate, the light is collected by an f/1 lens and focused on a photomultiplier tube. The absorption coefficient, b_{ab}, is determined from the Beer-Lambert Law in a way similar to that outlined for the integrating plate method.[4] This method measures the absorbing component of aerosol particles and is apparently insensitive to its scattering properties, as demonstrated by a wide range of experiments, including solvent extraction—thermal analysis studies[5] and photoacoustic spectroscopy.[6] (Also see results on pure $(NH_4)_2SO_4$ + mixtures of soot and $(NH_4)_2SO_4$ presented in this report.) Furthermore, Raman scattering[1] and thermal analysis experiments[7] indicate that the optical attenuation measurement is proportional to the graphitic content of aerosols collected directly from combustion sources or urban air.

The most direct substantiation that the optical attenuation technique selectively measures the absorbing component of aerosol particles is provided by a set of photoacoustic measurements[5] done in collaboration with the Applied Laser Spectroscopy group headed by Dr. N. M. Amer at LBL.

Unlike conventional optical absorption techniques, photoacoustic spectroscopy measures the energy deposited in a sample due to absorption. Since

questions have been raised whether the LBL laser transmission method exclusively measures the absorbing rather than the scattering component of the aerosol, a comparison between photoacoustic and optical attenuation measurements made on the same aerosol sample should help resolve this ambiguity.

The photoacoustic measurements were made in an acoustically nonresonant detector with cylindrical geometry (Fig. 2). A Knowles microphone (Model BT-1759 was used, and the cell dimensions were 2.1 cm in diameter and 0.3 cm in length. The gas in the detector cell was air at atmospheric pressure. An He-Ne laser operating at 632.8 nm with 0.5 mW of power was used as the light source, and the experiments were performed at a modulation frequency of 20 Hz. The aerosol particles, collected on 1.2-μm Millipore filter substrates, were mounted on a 1.5-mm-thick Pyrex backing with the particles facing the incident light beam. Experiments were also performed with the laser beam first incident on the filter substrate.

It is easy to show in the limit of low frequency light modulation[8] ($\leq$ 100 Hz) that the ratio of the photoacoustic signal to a reference sample for which the signal is saturated is given by:

$$S_{ph} = V/V_{sat} = 1 - \exp(-\alpha\ell) \ .$$

This saturable behavior was observed for highly absorbing samples, and the sample which yielded the largest photoacoustic signal was used as the reference, V_{sat}. Note that such samples yield values of $\alpha\ell \geq 3$, as deduced from the optical attenuation measurements; hence the highest signal obtained from available samples is close to the actual saturation value.

The experimental setup for the optical attenuation measurements is described above. In this technique the signal S_{op} is defined as $1 - \exp(-x)$, where x is the optical attenuation of the sample and is given by $-\ln I/I_0$,

where I is the transmitted intensity of a loaded filter and I_0 is the transmitted intensity of a blank filter.

In Fig. 3 we present a plot of the normalized photoacoustic signal S_{ph} vs. S_{op} for a wide range of ambient samples and samples collected directly from combustion sources. The samples include urban particulates collected over a 24-hr period in Fremont and Anaheim, California; Denver, Colorado; and New York, New York; and particles collected in a highway tunnel and from an acetylene torch. The least squares fit of the experimental points yields a correlation coefficient r of 0.98 and a slope of 1.03, which would be expected if both techniques measure the same optical property of the aerosol particles. Since the photoacoustic signal is proportional to the heat generated by absorption, we conclude that the optical-attenuation method measures the light-absorbing component of the aerosol particles.

From a theoretical point of view, this result is somewhat surprising, since aerosol particles have a large scattering coefficient, which would be expected to contribute to the optical attenuation measurement and not to the photoacoustic signal. This is especially true where the absorbing component represents only a small fraction of the aerosol mass. In this paper a simple model calculation will be presented which explains 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 of Lin et al.[4]

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. 4. 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 an almost 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.

$$I = I_0(1 - n_s\sigma_b - n_A\sigma_A) + I_0(1 - n_s\sigma_B - n_A\sigma_A)n_s\sigma_B R_F \tag{1}$$

$$+ I_0(1 - n_s\sigma_B - n_A\sigma_A)n_s^2\sigma_B^2 R_F^2 + \ldots I_0(1 - n_S\sigma_B - n_A\sigma_A)n_s^n\sigma_B^n R_F^n$$

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

Consider several limits. Where $R_F \approx 0$, 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, $R_F \approx 1$ and Eq. (1) becomes

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

Or the optical attenuation in the low loading limit is

$$ATN = I_0 - I = \frac{I_0 n_A \sigma_A}{1 - n_s\sigma_b} .$$

From this expression it is clear that a nonabsorbing aerosol will make no contribution 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, and if the scattering cross section of the particles, σ_s, is twice the particle area, then $n_s\sigma_s \approx 1$. If σ_B is about 20% of σ_s (the maximum value measured by Charlson and his coworkers[9]), then

$$1 - n_s\sigma_B \approx .8 ;$$

so that even for this 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 the LBL method for determining absorption coefficients and clearly is approximate since it assumes that the scattering properties of the particles are not affected by the filter substrate and neglects the penetration of the particles into the substrate. Future analysis will try to evaluate the significance of these effects.

Results of the Intercomparison

For the purpose of this intercomparison, parallel filter samples were collected on 47-mm Millipore (1.2-μm nominal pore size, type RATF) and prefired quartz fiber filters (Pallflex type 2500) at a nominal flow rate of .5 cfm. The flow rate through the filters was monitored continuously by a pressure sensor at the inlet of the pumps, and absolute flow rate calibrations were done on a daily basis using a Rockwell Model 5110 gas meter. (No altitude or temperature corrections were made in the gas meter flow rate calibration.) Optical attenuation measurements at a wavelength of .63 μ were done on both the Millipore and quartz fiber substrates. The optical attenuation is defined as ATN = -100 ln I/I_0, where I_0 is the intensity of the light transmitted through a blank filter substrate and I is the intensity through a loaded filter. The quartz filters were also used to determine the total carbon content of the aerosols by a combustion method.[10] Typical loading of dynamic blanks was 1 ± .3 $\mu g/cm^2$ of total carbon.

The optical attenuation measurements and total carbon loadings are reported in Tables I and II for the various aerosols investigated in the intercomparison. The optical attenuation measurements on quartz and Millipore substrates show a high degree of correlation as shown in Fig. 5. The least squares fit of the data has a correlation coefficient r = .99 and a slope of 1.15. The differences in the optical attenuation measurement on these two rather different collection media appear to be minimal. The optical attenuation measurements also show a high degree of correlation with the total carbon content of the combustion-generated aerosols (soot and soot + sulfate) as shown in Fig. 6. Such a correlation would be expected when graphitic carbon represents a fixed fraction of the total carbon content of the aerosol. No systematic differences are found in the slope of this line, even when considerable amounts of $(NH_4)_2SO_4$ are

present. Although this data set is limited, the result is a further confirmation of the fact that the optical attenuation measurement is insensitive to the scattering properties of the aerosol. It should also be noted that the optical attenuation of a pure $(NH_4)_2SO_4$ aerosol was below the detection limit of the measurement (see Table I).

The absorption coefficients reported in Table I were obtained from optical attenuation measurements on the Millipore substrates in the following manner:

$$b_{ab}(m^{-1}) = \frac{(ATN)(10^{-6})C}{F},$$

where F is the flow rate in m^3/cm^2 and C is an experimentally determined correction factor which takes into account the penetration of particles in the filter substrate. For the data reported here, a correction factor of .62 has been used. This correction term has been determined in a preliminary way by comparing the optical attenuation of parallel filter samples of urban air collected on Millipore and very thin, almost transparent Teflon substrates where enhanced absorption due to multiple scattering is expected to be small (Fig. 7). The optical attenuation measurement on the Teflon filters was made with the geometry shown in Fig. 4, using a blank Millipore filter as the diffuse reflector. Also included in Table I are values of the specific absorption coefficient for the soot aerosols. These values are given whenever the carbon loading is determined with an accuracy of ±25% or better. The absorption coefficients reported here are consistent with the optical constnats of graphitic carbon and are expected to have an accuracy of better than a factor of two.

Acknowledgment

This work was supported by the Assistant Secretary for the Environment, Office of Health and Environmental Research, Pollutant Characterization and Safety Division of the U.S. Department of Energy under Contract No. W-7405-ENG-48 and by the National Science Foundation under Grant No. ATM 80-13707.

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Table I. Optical attenuation measurements, absorption coefficients, and specific absorption coefficients determined from Millipore substrates.

Sample	Date	Time	Aerosol	Flow (m^3/cm^2)	ATN	b_{ab} (m^{-1})	μg C/m^3 [a]	Specific absorption coefficient (m^2/g)
7/29/1	7/29	1529-1600	$(NH_4)_2SO_4$	$5.0x10^{-2}$	0	b		
7/29/2	7/29	1635-1733	$(NH4)_2SO_4$ + 1% MB	$9.4x10^{-2}$	52	$3.4x10^{-4}$		
7/30/1	7/30	1120-1232	$(NH_4)_2SO_4$	$1.08x10^{-1}$	-4.8	a		
7/30/2	7/30	1327-1527	$(NH_4)_2SO_4$ + .3% MB	$1.8x10^{-1}$	8.6	$3.0x10^{-5}$		
7/30/3	7/30	1636-1741	$(NH_4)_2SO_4$ + 4% MB	$9.8x10^{-2}$	85	$5.4x10^{-4}$		
7/31/1	7/31	1015-1025	Soot	$1.4x10^{-2}$	44	$1.9x10^{-3}$	193	9.8
7/31/2	7/31	1035-1040	Soot	$6.8x10^{-3}$	21	$1.9x10^{-3}$		
7/31/3	7/31	1048-1113	Soot	$3.5x10^{-2}$	79	$1.4x10^{-3}$	180	7.8
7/31/4	7/31	1124-1126	Soot	Abort				
7/31/5	7/31	1133-1136	Soot	$4.2x10^{-3}$	14	$2.1x10^{-3}$		
7/31/6	7/31	1143-1145	Soot	$2.4x10^{-3}$	3.4	—		
7/31/7	7/31	1157-1204	Soot	$1.0x10^{-2}$	35	$2.2x10^{-3}$		
7/31/8	7/31	1215-1230	Soot	$2.1x10^{-2}$	65	$1.9x10^{-3}$	177	10.7
7/31/9	7/31	1238-1345	Soot	$9.5x10^{-2}$	163	$1.1x10^{-3}$	159	6.9
7/31/10	7/31	1442-1452	Soot + $(NH_4)_2SO_4$	$1.4x10^{-2}$	18	$8.0x10^{-4}$		
7/31/11	7/31	1502-1522	Soot + $(NH_4)_2SO_4$	$2.8x10^{-2}$	35	$7.8x10^{-4}$		
7/31/12	7/31	1533-1640	Soot + $(NH_4)_2SO_4$	$9.5x10^{-2}$	86	$5.6x10^{-4}$	48	11.7
8/1/1	8/1	1130-1203	MB	$3.9x10^{-2}$	118	$1.9x10^{-3}$		
8/1/2	8/1	1212-1218	MB	$7.8x10^{-3}$	36	$2.9x10^{-3}$		
8/1/3	8/1	1226-1239	MB	$1.7x10^{-2}$	85	$3.1x10^{-3}$		
8/1/4	8/1	1247-1325	MB	$4.9x10^{-2}$	133	$1.7x10^{-3}$		
8/1/5	8/1	1435-1458	Soot	$2.8x10^{-2}$	68	$1.5x10^{-3}$		
8/1/6	8/1	1505-1510	Soot	$6.6x10^{-3}$	18	$1.7x10^{-3}$		
8/1/7	8/1	1517-1527	Soot	$1.3x10^{-2}$	31	$1.5x10^{-3}$		
8/4/1	8/4	1053-1113	Soot	$2.8x10^{-2}$	69	$1.5x10^{-3}$		
8/4/2	8/4	1140-1208	Soot + $(NH_4)_2SO_4$	$3.9x10^{-2}$	85	$1.4x10^{-3}$		
8/4/3	8/4	1214-1219	Soot + $(NH_4)_2SO_4$	$6.6x10^{-3}$	28	$2.6x10^{-3}$		
8/4/4	8/4	1226-1315	Soot + $(NH_4)_2SO_4$	$6.5x10^{-2}$	99	$9.4x10^{-4}$		
8/4/5	8/4	1322-1332	Soot + $(NH_4)_2SO_4$	$1.3x10^{-2}$	37	$1.8x10^{-3}$		
8/4/6	8/4	1337-1352	Soot + $(NH_4)_2SO_4$	$2.0x10^{-2}$	47	$1.5x10^{-3}$		
8/5/1	8/5-8/6	0027-0827	Ambient	1.7	22	$8x10^{-6}$		
8/6/1	8/6	1044-1331	Arizona	$2.1x10^{-1}$	4.1	a		
8/6/2	8/6	1455-1659	Soot + $(NH_4)_2SO_4$	$1.6x10^{-1}$	113	$4.4x10^{-4}$		
8/6/3	8/6	1741-1802	Soot	$2.7x10^{-2}$	83	$1.9x10^{-3}$	190	10.0
8/7/1	8/7	1145-1224	Soot + $(NH_4)_2SO_4$	$4.9x10^{-2}$	50	$6.3x10^{-4}$		Average specific absorption = 9.5 ± 1.8 m^2/g
8/8/1	8/8	0952-1051	MB	$7.5x10^{-2}$	219	$1.8x10^{-3}$		

[a] Only total carbon loadings with an error of less than ±25% are reported.

[b] Below detection limit.

Table II. Optical attenuation measurements and total carbon loadings determined from quartz substrates.

Sample	Date	Time	Aerosol	Flow (m^3/cm^2)	ATN	ATN[a]	$\frac{\mu g\ C}{cm^2}$	$\frac{\mu g\ C}{m^3}$
7/29/1	7/29	1529-1600	$(NH_4)_2SO_4$	4.8×10^{-2}	1.7	b		
7/29/2	7/29	1635-1733	$(NH_4)_2SO_4$ + 1% MB	8.7×10^{-2}	57	62		
7/30/1	7/30	1120-1232	$(NH_4)_2SO_4$	1.0×10^{-1}	-2	b		
7/30/2	7/30	1327-1526	$(NH_4)_2SO_4$ + .3% MB	1.7×10^{-1}	12	13		
7/30/3	7/30	1636-1741	$(NH_4)_2SO_4$ + 4% MB	9.4×10^{-2}	93	97		
7/31/1	7/31	1015-1025	Soot	1.4×10^{-2}	57	57	2.7	193
7/31/2	7/31	1035-1040	Soot	7.3×10^{-3}	32	30		
7/31/3	7/31	1048-1113	Soot	3.6×10^{-2}	97	94	6.5	180
7/31/4	7/31	1124-1126	Soot	2.9×10^{-3}	9.3			
7/31/5	7/31	1133-1136	Soot	4.4×10^{-3}	18	17		
7/31/6	7/31	1143-1145	Soot	2.5×10^{-3}	6.4	6.1		
7/31/7	7/31	1157-1204	Soot	1.1×10^{-2}	46	42		
7/31/8	7/31	1215-1230	Soot	2.2×10^{-2}	76	73	3.9	177
7/31/9	7/31	1238-1345	Soot	9.7×10^{-2}	206	202	15.4	159
7/31/10	7/31	1442-1452	Soot + $(NH_4)_2SO_4$	1.3×10^{-2}	18	19		
7/31/11	7/31	1502-1522	Soot + $(NH_4)_2SO_4$	2.9×10^{-2}	41	39		
7/31/12	7/31	1533-1640	Soot + $(NH_4)_2SO_4$	9.7×10^{-2}	100	98	4.7	48
8/1/1	8/1	1130-1203	MB	4.1×10^{-2}	185	176		
8/1/2	8/1	1212-1218	MB	9.0×10^{-3}	114	99		
8/1/3	8/1	1226-1238	MB	1.8×10^{-2}	153	145		
8/1/4	8/1	1247-1325	MB	5.8×10^{-2}	169	143		
8/1/5	8/1	1435-1458	Soot	3.4×10^{-2}	79	65		
8/1/6	8/1	1505-1510	Soot	7.5×10^{-3}	19	17		
8/1/7	8/1	1517-1527	Soot	1.5×10^{-2}	41	36		
8/6/3	8/6	1741-1802		3.2×10^{-2}	116	98	6.1	190
8/7/1	8/7	1145-1224		5.9×10^{-2}	41	34		
8/8/1	8/8	0952-1051		8.9×10^{-2}	310	261		
8/6/2(?)	8/6	1455-1659	Soot + $(NH_4)_2SO_4$	.19	140	118		

[a]Normalized to Millipore flowrate

[b]Below detection limit

Figure Captions

Figure 1. Schematic of LBL laser transmission apparatus.

Figure 2. Photoacoustic experimental arrangement.

Figure 3. Plot of S_{ph} vs. S_{op} for various samples:

▽ - Fremont: □ - Anaheim; ○ - Denver; △ - New York City;

■ - highway tunnel; ● - acetylene torch.

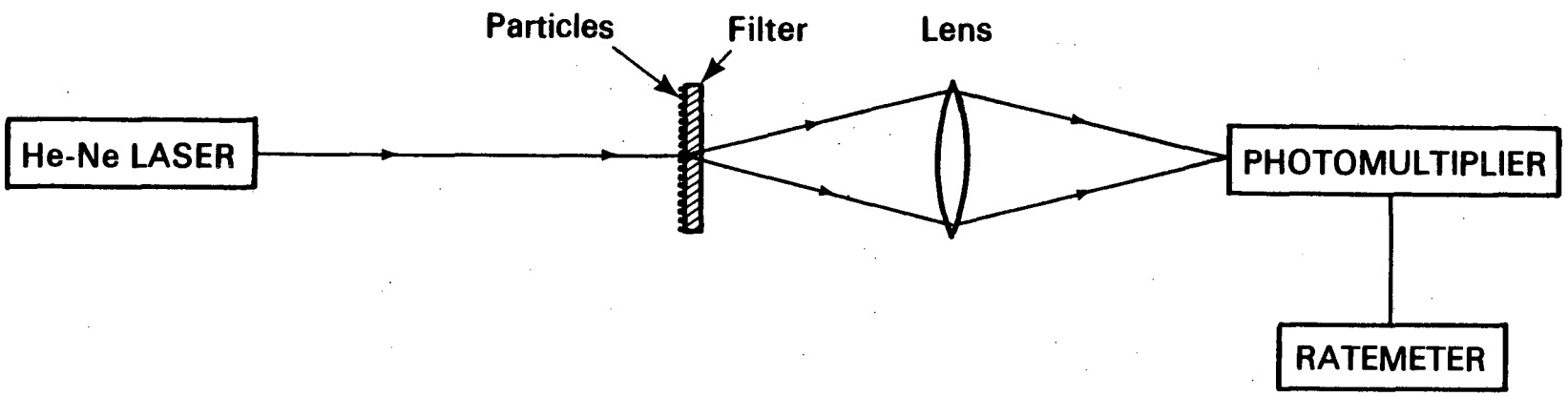
The solid line is a least squares fit of the data.

Figure 4. Schematic of experimental arrangement used for model calculation.

Figure 5. Comparison of optical attenuation determined on quartz and Millipore substrates.

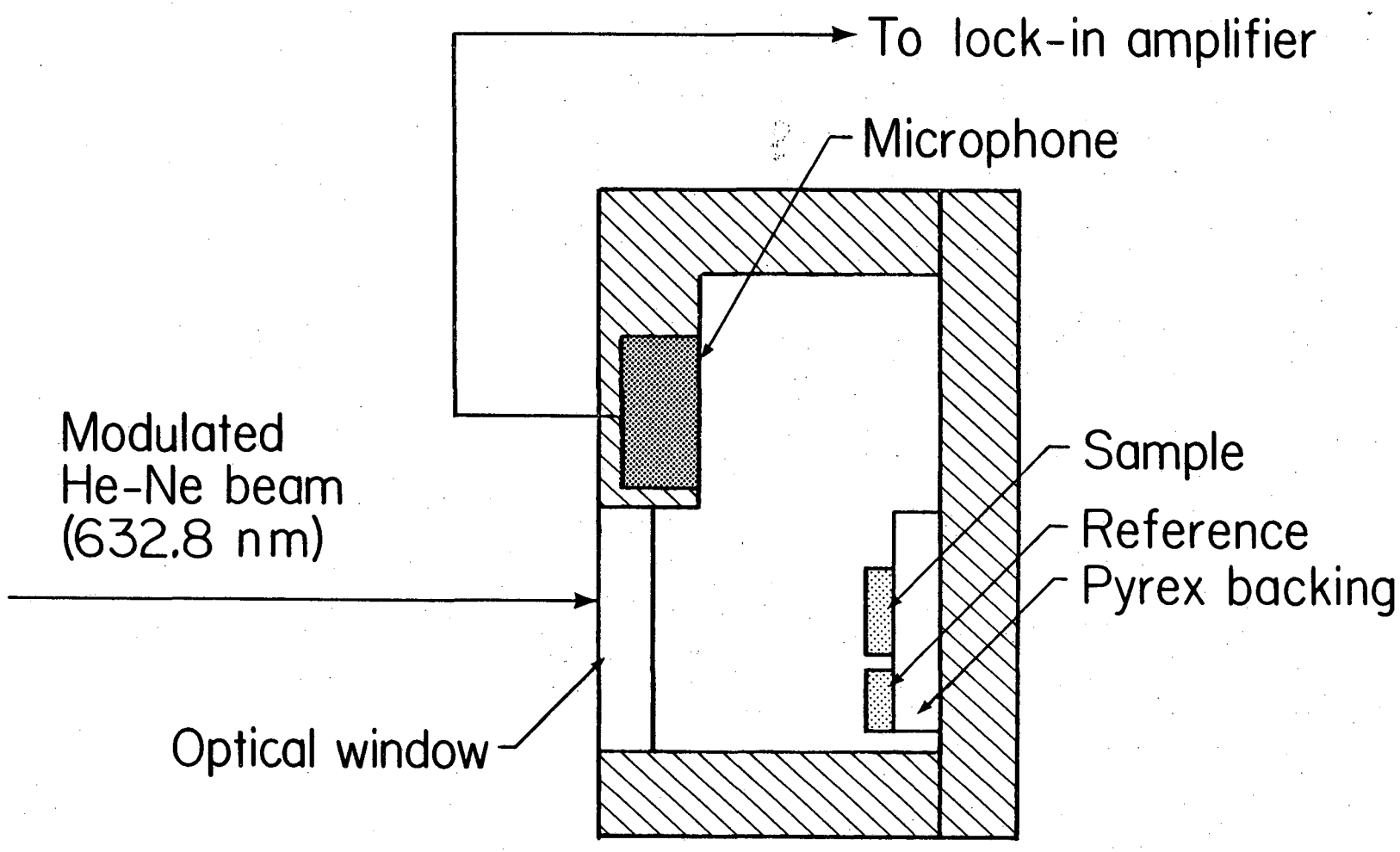
Figure 6. Comparison of optical attenuation determined on quartz substrates with total carbon loading for combustion-generated aerosols.

Figure 7. Comparison between optical attenuation measurements on Millipore and Teflon substrates.



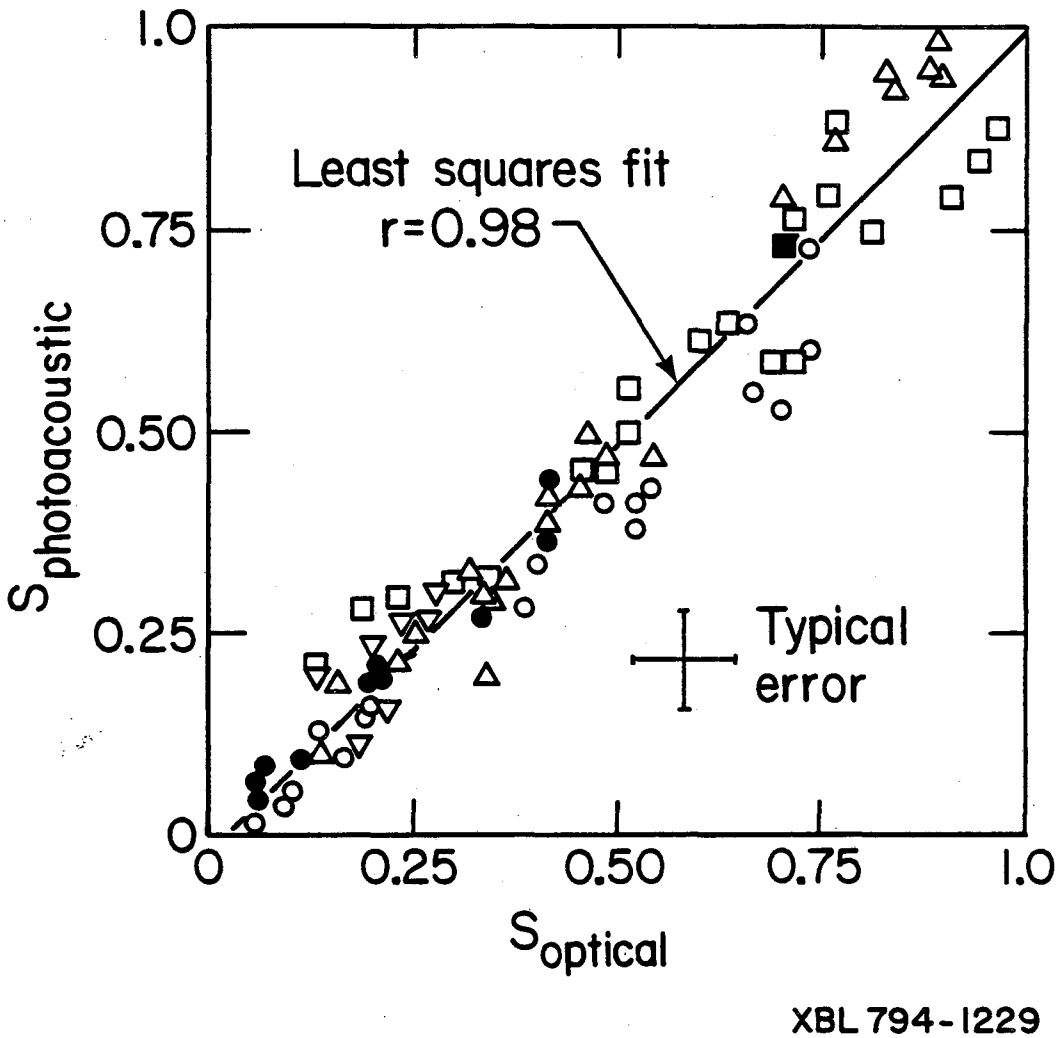
XBL 787-1361

Figure 1



XBL 794-1230

Figure 2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Least squares fit
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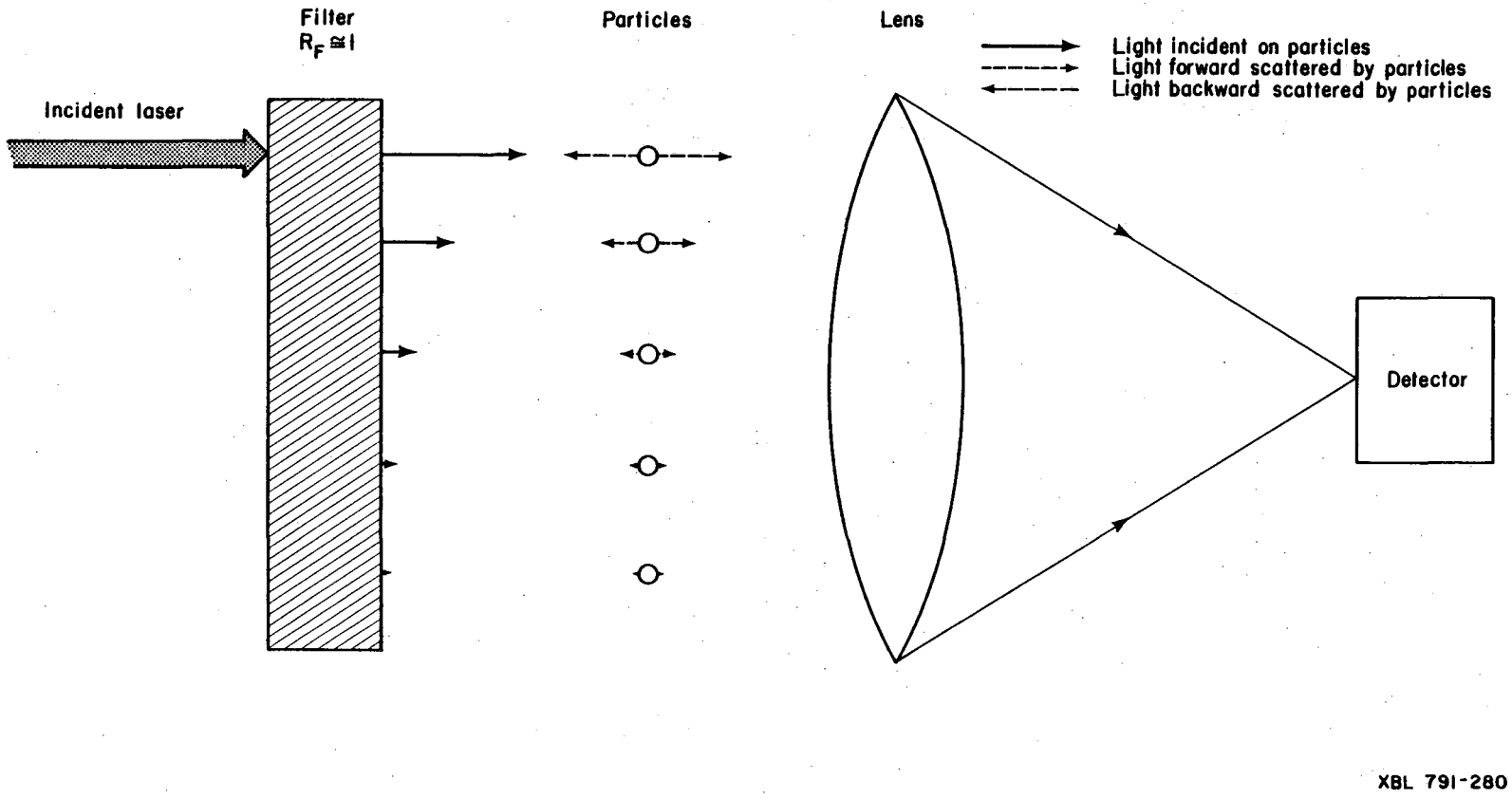
Typical
error
$S_{photoacoustic}$
$S_{optical}$
XBL 794-1229

Figure 3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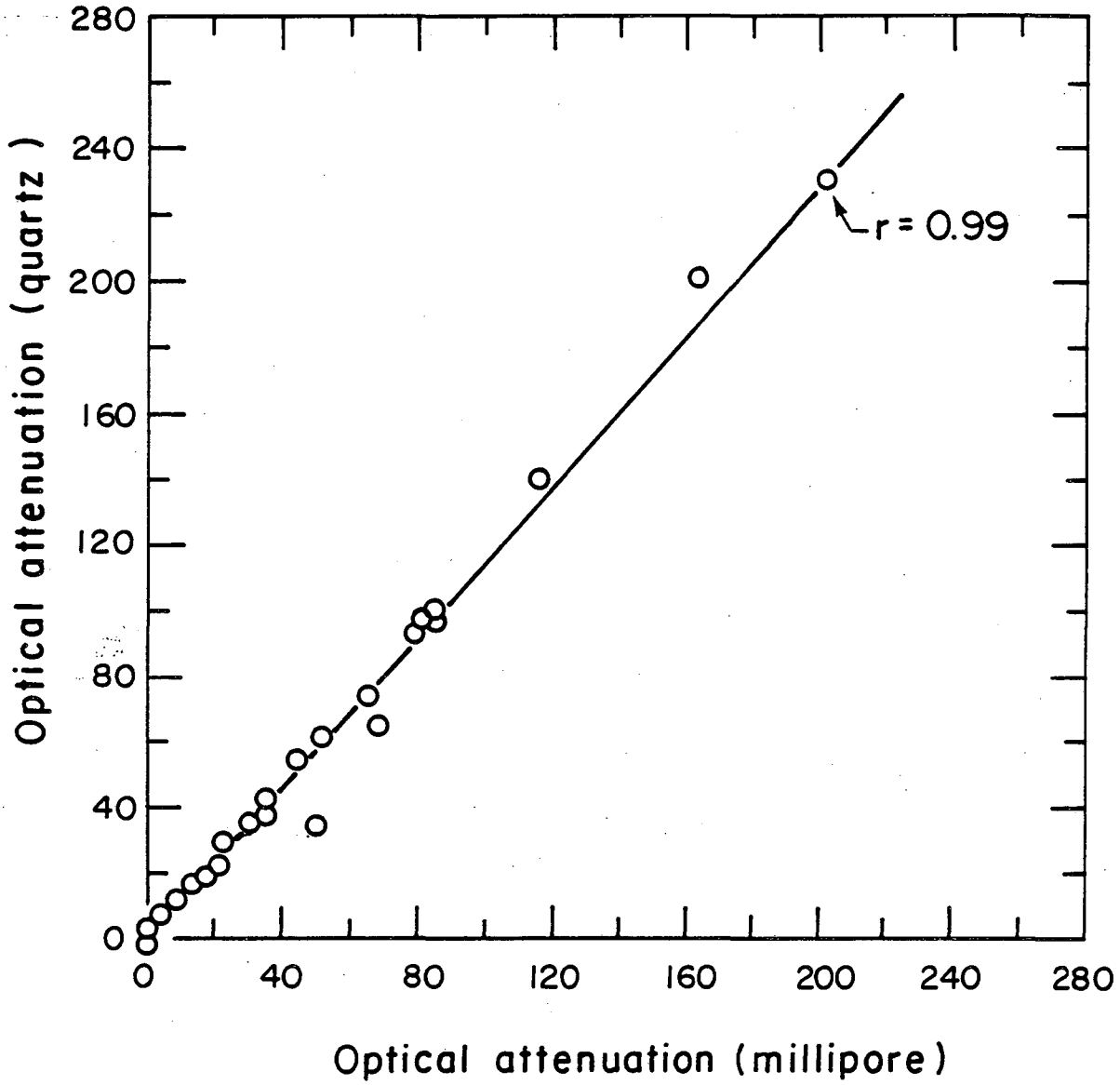
$R_F \cong 1$
Particles
Lens
Light incident on particles
Light forward scattered by particles
Light backward scattered by particles
Incident laser
Detector
XBL 791-280

Figure 4



XBL 817-2408

Figure 5

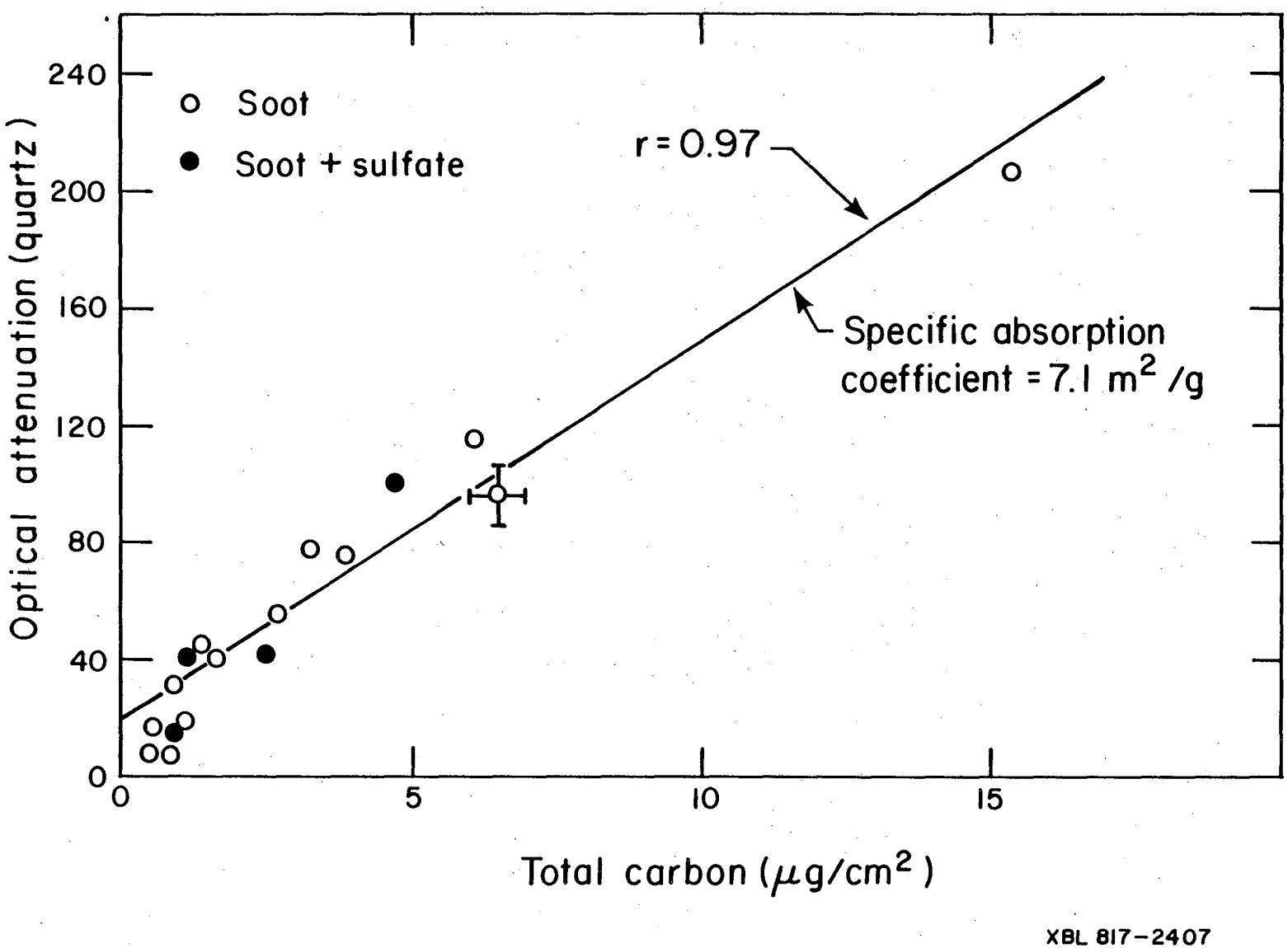
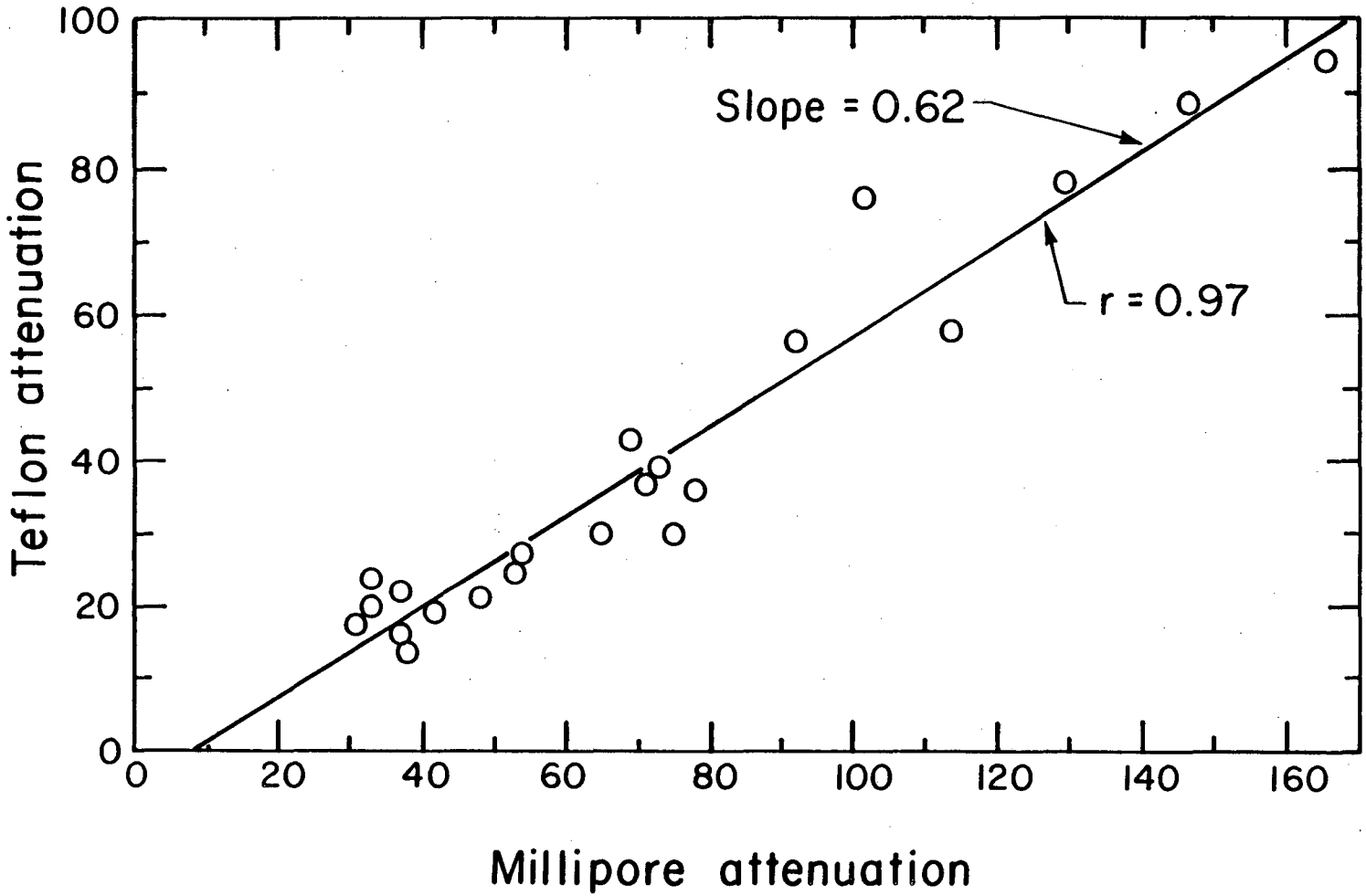

Soot
Soot + sulfate
r = 0.97
Specific absorption coefficient = 7.1 m² /g
Optical attenuation (quartz)
Total carbon (μg/cm²)
0
40
80
120
160
200
240
0
5
10
15
XBL 817-2407

Figure 6



XBL 817-2415

Figure 7

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TECHNICAL INFORMATION DEPARTMENT
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720