

COMPARISON BETWEEN THE LIGHT EXTINCTION AEROSOL MASS CONCENTRATION RELATIONSHIP OF ATMOSPHERIC AND AIR POLLUTANT EMISSION AEROSOLS

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Abstract—A comparison between the measured and calculated light extinction–aerosol mass concentration relationship for atmospheric and source emission aerosols is presented. A parameter K , defined as the specific particulate volume/light extinction coefficient ratio ($\text{cm}^3 \text{m}^{-3}/\text{m}^{-1}$) is used in the comparison. The measured magnitudes range from 0.256 to 0.487 for averaged atmospheric K s and from 0.06 to 0.78 for individual source emission K s. Additional K data for source emissions and background atmospheric aerosols would be useful for estimating the contribution of various aerosol sources to atmospheric visibility reduction.

INTRODUCTION

THE DETECTION and evaluation of particulate air pollutants, both in the general atmosphere and in smoke or dust plumes, is most frequently performed by visual observation. Relationships between the mass concentration of particulate matter and light extinction (atmospheric visibility or plume opacity) have been reported for the atmosphere by STEFFEN (1956), CHARLSON *et al.* (1967), and NOLL *et al.* (1968), for particulate air pollutant sources in general by PILAT and ENSOR (1970), and for specific particle size distributions by HAWKSLEY *et al.* (1961) and by ROBINSON (1962). A comparison between the light extinction–aerosol mass concentration relationship for the atmospheric aerosol and for specific particulate emissions is of significance because:

1. The results should assist in the development of air resource management programs for improving the visual quality of air by providing some insight concerning the effect of source emissions on atmospheric visibility.
2. The comparison may imply whether the atmospheric aerosol size distribution is influenced more by emission source particle size distributions or by events occurring in the atmosphere (coagulation, sedimentation, condensation, chemical reaction, etc.).

DEVELOPMENT OF EQUATIONS RELATING LIGHT EXTINCTION TO AEROSOL PROPERTIES

A general relationship can be developed between the light transmittance (I/I_0), the illumination or visual path length L , and the particle properties (particle size distribution, density, mass concentration, and refractive index). The transmission of light through a volume containing an aerosol is described by the Lambert–Beer law.

$$I/I_0 = \exp(-B_E L) \quad (1)$$

Where L is the illumination path length and B_E is the light extinction coefficient. Assuming the light extinction is due only to aerosol particles, B_E can be defined in

terms of the extinction efficiency factor Q_E and the particle number frequency size distribution $n(r)$.

$$B_E = \int_0^{\infty} Q_E(r, \lambda, m) \pi r^2 n(r) dr. \quad (2)$$

The particle mass concentration W is related to $n(r)$ and the particle density ρ by

$$W = \int_0^{\infty} \frac{4}{3} \pi r^3 \rho n(r) dr. \quad (3)$$

PILAT and ENSOR (1970) and CHARLSON *et al.* (1967) used a parameter K to combine the variable quantities of the aerosol size distribution $n(r)$ and the light extinction efficiency factor Q_E . K is defined as the specific particulate volume (cm^3 particles m^{-3} air) divided by the light extinction coefficient (m^{-1}) and can be referred to as the (specific particulate volume)/(light extinction coefficient) ratio ($\text{cm}^3 \text{m}^{-2}$). K is defined mathematically as

$$K = \frac{\frac{4}{3} \int_0^{\infty} r^3 n(r) dr}{\int_0^{\infty} r^2 Q(r, \lambda, m) n(r) dr}. \quad (4)$$

The relationship of the parameter K to the other variables can be shown by an equation of the form of the Lambert-Beer law

$$\ln \frac{I}{I_0} = - \frac{WL}{K\rho} \quad (5)$$

or of a form relating K to the light extinction coefficient.

$$K = \frac{W}{B_E \rho}. \quad (6)$$

CHARLSON *et al.* (1968) presented a relationship between the atmospheric aerosol concentration and light extinction using meteorological visual range considerations. The Koschmieder visibility theory relates meteorological range L_v to the light extinction coefficient B_E at a wavelength of $550 \mu\text{m}$ by

$$L_v = \frac{3.9}{B_E}. \quad (7)$$

Multiplying the above equation by the aerosol mass concentration W gives

$$L_v W = \frac{3.9 W}{B_E}. \quad (8)$$

Substituting equation (6) for W/B_E in equation (8) provides a relationship between meteorological range L_v and K .

$$L_v W = 3.9 K\rho \quad (9)$$

solving for K

$$K = \frac{L_v W}{3.9\rho}. \quad (10)$$

Thus the atmospheric K may be determined by either human observations or instrumental measurements of visibility combined with measurements of the atmospheric aerosol mass concentration W .

COMPARISON BETWEEN THE PARTICULATE VOLUME/EXTINCTION COEFFICIENT RATIO OF THE ATMOSPHERE AND FOR EMISSION SOURCES

A. Magnitude of atmospheric K

Measurements of the atmospheric K have been reported by CHARLSON *et al.* (1967) and NOLL *et al.* (1968). CHARLSON *et al.* (1968) reported that the most probable value of K (assuming aerosol particles of unit density) is 0.308 with 90 per cent of all measurements ranging between 0.18 and 0.67. NOLL *et al.* (1968) reported data which has been converted into the parameter K and is listed in TABLE 1.

TABLE 1. MEASURED ATMOSPHERIC PARTICULATE VOLUME/EXTINCTION COEFFICIENT RATIOS

Study	I/I_0 measurement	Particle mass measurement	K ($\text{cm}^3 \text{m}^{-2}$)
Berkeley-Oakland	Human observer	Filtration, 2.5 cm dia. glass fiber filter	0.487
Bell at Berkeley	Volz sun-photometer	High-volume filter	0.411
Soiling index	Soiling index	High-volume filter	0.257
Charlson at Seattle	Nephelometer	Filtration, 2.5 cm dia. glass fiber filter	0.308

Calculated magnitudes of K have been reported by CHARLSON *et al.* (1968) which varied from 0.22 to 0.95 m^{-2} for aerosol size distribution slopes ranging from 2 to 7 as shown in TABLE 2.

TABLE 2. CALCULATED ATMOSPHERIC PARTICULATE VOLUME/EXTINCTION COEFFICIENT RATIO

β	2	3	4	5	6	7
$K(\text{cm}^3 \text{m}^{-2})$	0.56	0.33	0.22	0.30	0.60	0.95

Their calculations included assumptions of a particle refractive index of 1.5, a wavelength of 4700 Å, a particle number frequency distribution $n(r) = Ar^{-\beta}$ with upper and lower radius limits of 2 and $0.04 \mu\text{m}$, respectively.

B. Magnitudes of particulate emission source K

Simultaneous measurements of the aerosol mass concentration, the plume light transmittance, and the stack diameter at source emissions have been reported by

STOECKER (1950), HURLEY and BAILEY (1958), and CONNER and HODKINSON (1967). A summary of the published data and new data obtained during this study by ENSOR and PILAT (1969) and by BOSCH (1969) is presented in TABLE 3.

TABLE 3. MEASURED PARTICULATE VOLUME/EXTINCTION COEFFICIENT RATIO OF EMISSION SOURCES

Study	Emission	I/I_0 measurement	Particle mass measurement	K ($\text{cm}^3 \text{m}^{-2}$)
STOECKER (1950)	Black smoke (coal)	Light-photocell	Alundum filter	0.11
HURLEY and BAILEY (1958)	Black smoke (coal)	Light-photocell	Filtration	0.084
CONNER and HODKINSON (1967)	White smoke	Transmissometer	Membrane filters	0.32
CONNER and HODKINSON (1967)	Black smoke	Transmissometer	Membrane filters	0.059
ENSOR and PILAT (1969)	Coal fly ash	Bailey smoke meter	Alundum filter	0.78
BOSCH (1969)	Kraft recovery furnace	Bailey smoke meter	Alundum filter	0.68

PILAT and ENSOR (1970) reported calculated magnitudes of the parameter K for log-normal aerosol size distributions as a function of the geometric mass mean radius and the geometric standard deviation for spherical particles of refractive indices 1.33 and $1.95-0.66i$ at a wavelength of 5000 Å. The calculated K 's vary from 0.05 to 70 for mass mean radii in the range from 0.1 to 100 μm and standard deviation from 1.0 to 5. The major variation in the parameter K is caused by changes in the particle size distribution. The calculated magnitudes of K as a function of mass mean radius and geometric standard deviation are presented in TABLES 4-6 for refractive indices 1.33, 1.5, and $1.95-0.66i$, respectively.

TABLE 4. CALCULATED K FOR 1.33 REFRACTIVE INDEX

σ	0.1	0.5	1.0	\bar{r}_{gw}	5	10	50	100
1	0.67	0.17	0.70		3.02	6.67	33.3	66.7
2	0.47	0.24	0.39		2.39	5.01	26.2	52.4
3	0.45	0.30	0.39		1.55	3.25	17.8	36.2
4	0.47	0.35	0.42		1.19	2.26	12.0	24.7
5	0.49	0.39	0.45		1.04	1.79	8.4	17.1

TABLE 5. CALCULATED K FOR 1.50 REFRACTIVE INDEX

σ	0.1	0.5	1.0	\bar{r}_{gw}	5	10	50	100
1	0.29	0.28	0.51		3.06	6.67	33.3	66.7
2	0.23	0.19	0.39		2.40	5.02	26.2	52.4
3	0.25	0.22	0.33		1.53	3.26	17.9	36.2
4	0.28	0.25	0.33		1.10	2.19	12.0	24.7
5	0.30	0.28	0.34		0.92	1.66	8.3	17.0

TABLE 6. CALCULATED K FOR 1.95-0.66i REFRACTIVE INDEX

σ	0.1	0.5	1.0	F_{sw}	5	10	50	100
1.	0.044	0.26	0.57		3.13	6.66	33.3	66.7
2	0.068	0.20	0.42		2.41	5.02	26.2	52.4
3	0.081	0.17	0.30		1.57	3.31	17.9	36.2
4	0.090	0.16	0.25		1.08	2.21	12.1	24.8
5	0.096	0.16	0.23		0.82	1.59	8.3	17.1

DISCUSSION OF RESULTS

A. Effect of aerosol source emissions on the atmospheric K

The examination of a simple visibility model may provide some insight concerning an approach for determining the effect of source emissions on the atmospheric K . Consider the atmosphere in an air basin as a well mixed box under steady state conditions. Natural ventilation transports background atmospheric aerosol into the box and polluted air out. Assuming no modifications of the source aerosols other than concentration dilution and considering the formation of aerosols by atmospheric reactions as another source, the atmospheric K may be given by

$$\frac{1}{K_{\text{Atm}}} = \frac{F_R}{K_{\text{Atm reactions}}} + \frac{F_V}{K_{\text{Air into basin}}} + \frac{F_S}{K_{\text{Emission sources}}} \quad (11)$$

where F_R is the ratio of the particle volume/time of aerosols formed by atmospheric reactions and vapor condensation to the volume/time of total aerosols injected into the air basin box. The particulate volume fraction F_R is a function of the atmospheric gaseous pollutant concentrations, sunlight intensity, temperature, etc. F_V is the volume fraction of particulate matter carried by winds into the air basin box and is dependent upon the wind speed and background aerosol concentration. F_S is the volume fraction of particulates introduced into the air basin box from aerosol emission sources and is given by

$$\frac{F_S}{K_{\text{Emission source}}} = \sum_{i=1}^N \frac{F_i}{K_i} \quad (12)$$

where F_i is the ratio of the volume/time of aerosols emitted into the air basin box by source i to the volume/time of particulates emitted into the atmosphere by the total number of aerosol sources N in the air basin. K_i is the specific particulate volume/light extinction coefficient ratio for source i . Although equation (11) does not provide for changes in the aerosol size distribution caused by coagulation or sedimentation, it does present a method for calculating the atmospheric K . Equation (11) may also be used to estimate the contribution of various aerosol sources to reductions in atmospheric visibility.

This preliminary analysis indicates that data is needed concerning the K 's and input rates of individual aerosol sources, aerosols from atmospheric reactions, and background atmospheric aerosols. Although emission source and atmospheric aerosol size distribution information would essentially serve the same purpose [calculate K by equation (4)], K data appears to be somewhat easier to obtain by simultaneous

measurements of the light extinction coefficient B_E and the aerosol mass concentration W [calculate K by equation (6)].

B. Relationship of K measurements to atmospheric aerosol size distribution theory

The magnitude of K is dependent upon the aerosol size distribution. Thus K measurements may provide some information concerning the observed shape of the atmospheric aerosol size distribution. For example the K for a parcel of aerosol can vary from the point of emission or formation until it becomes the atmospheric aerosol due to changes in the size distribution caused by coagulation and/or sedimentation. In general the magnitude of K should be increased by coagulation and decreased by sedimentation (assuming the geometric mass mean radius of the aerosol is greater than about 0.1–0.4 μm). However, no trends (increasing or decreasing) in K from an emission source to the atmospheric aerosol are indicated by the presently available data. The ranges in the individual measurements of the atmospheric K 's (0.18–0.67) and of the source emission K 's (0.059–0.78) are actually quite similar.

There are at least two basically different theories for the observed regular shape of the atmospheric aerosol size distribution $n(r)$:

1. That the atmospheric $n(r)$ is due to processes occurring in the atmosphere and is independent of the source $n(r)$, and
2. that the atmospheric $n(r)$ is the result of the statistical combination of the $n(r)$ of aerosol sources.

FRIEDLANDER (1960) proposed that the atmospheric aerosol size distribution above 0.1 μm radius is the result of processes (coagulation and sedimentation) occurring in the atmosphere. Later FRIEDLANDER (1963, 1965) developed the self-preserving size distribution theory based on similarity solutions to the equations for the rate of change of the aerosol size distribution with time. JUNGE (1965, 1969) has suggested that the shape of the atmospheric aerosol size distribution above 0.1 μm radius is a statistical effect of the combination of aerosols from emission sources. JUNGE (1955) also reported that the shape of the atmospheric aerosol size distribution above 0.1 μm radius cannot be modified to any degree by coagulation and thus the self-preserving size distribution theory (with coagulation) is unrealistic for the atmospheric aerosol. CLARK and WHITBY (1967) concluded that their measured atmospheric aerosol size distributions above 0.05 μm radius were in agreement with Friedlander's self-preserving size distribution theory. However, JUNGE (1969) reported that the test proposed by Friedlander and used by CLARK and WHITBY (1967) to verify the self-preserving size distribution theory is not valid. JUNGE (1969) maintains that the test is a necessary but not a sufficient condition and thus cannot be considered as proof of the self-preserving size distribution theory.

PILAT (1969) pointed out the need for more data concerning the particle size distributions of aerosol emission sources in order to test Junge's theory for the atmospheric aerosol size distribution. Additional measurements of atmospheric and aerosol emission source K 's may also provide some useful information about the observed regular shape of the atmospheric aerosol size distribution. However, it should be noted that equal atmospheric and emission source K 's is not conclusive proof of equal aerosol size distributions as each magnitude of K does not have a unique aerosol size distribution (there is a minimum K for both the Junge size distribution as shown in TABLE 2 and for the log-normal size distribution as shown in TABLES 4–6).

CONCLUSIONS

The measured magnitudes of K range from 0.256 to 0.487 for the atmosphere (averaged at given locations) and from 0.059 to 0.78 for individual source emission aerosols. Additional data concerning the K 's and rates of input into the atmosphere of individual emission source aerosols, of aerosols formed by atmospheric reactions, and of background atmospheric aerosols would be useful for estimating the contribution of various aerosol sources to atmospheric visibility reduction. This data may also provide some information concerning the observed regular shape of the atmospheric aerosol size distribution.

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