Calculation of Smoke Plume Opacity from Particulate Air Pollutant Properties

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Calculation of smoke plume opacity from the properties of the particulate emission is facilitated with the use of a parameter K (specific particulate volume cm^3/m^3/extinction coefficient m^{-1}) computed from theory. Graphs of K vs. the geometric mass mean particle radius at geometric standard deviations from 1 (monodisperse) to 10 are presented for particle refractive indices of 1.96–0.66i (carbon), 2.80–0.02i, 1.33 (water) and 1.50 at a wavelength of light of 550 nm. Experimental data of K for various sources are reported. Application to the estimation of the Ringelmann number is discussed and illustrated with an example.

Nomenclature

- b — extinction coefficient per volume of aerosol
- (r) — particle number fraction frequency distribution
- I — intensity of transmitted light
- I_0 — intensity of incident light
- I/I_0 — light transmittance
- i — $(-1)^{1/2}$
- K — specific particulate volume/extinction coefficient ratio
- L — illumination path length or diameter of plume
- m — refractive index of particle relative to air
- n_1 — real part of refractive index
- n_2 — imaginary part of refractive index
- $n(r)$ — particle number frequency distribution, which, multiplied by the radius increment, dr, is the number of particles between r and $r + dr$
- Q_E — extinction efficiency factor
- r — particle radius
- r_{gn} — geometric number mean radius
- r_{gw} — geometric mass mean radius
- Sp — specific projected area or the specific extinction area
- W — total particulate mass concentration

Greek Symbols

- α — size parameter, $2\pi r/\lambda$
- λ — wavelength of light
- π — 3.14159
- ρ — average particle density
- σ_g — particle size geometric standard deviation

Introduction

Air Pollution Regulations

Regulations to control particulate air pollution from sources are usually of the following types:

1. visible emissions (Ringelmann No.);
2. particulate matter concentration grain loading (grains/scf);
3. process weight code (lb particu'ate emission/lb process weight);
4. boundary line atmospheric aerosol concentrations (micrograms/cubic meter).

A detailed discussion of the various air pollution regulations and the communities implementing them can be found in Stern.[1] The regulation of sources by visible evaluation of the emission plume opacity (Ringelmann No. limit) is the least time consuming and least expensive method for a control agency to implement. For these reasons Ringelmann number observations are frequently used to control sources. In Table I the Ringelmann number, plume opacity, and fractional light transmittance are compared. Ringelmann numbers are used by an observer to describe black smoke plumes, and equivalent opacity is used for white or colored plumes.

A relationship between the plume opacity and the particle physical properties (especially mass concentration) would aid in the design of control equipment and indicate correspondence between the various types of regulations for particulate air pollutants. Often an emission source will meet the particulate matter concentration requirement but not the visual standard (plume opacity).

Previous Work

Correlations of particle mass concentration to light transmittance have been reported for a few air pollution sources. Stoecher[2] and Hurley and Bailey[3] reported measurements of particulate mass concentration and opacity from coal combustion. Stern[1] reported the Ringelmann number expected for various fly ash concentrations from a coal fired power plant. Simultaneous measurements of particulate mass concentration and plume transmittance for white and black (fuel oil) smoke sources were reported by Conner and Hodkinson.[4] Gansler[5] reported smoke meter measurements of stack transmittance and mass concentration for the emissions from a Kraft recovery furnace. These studies indicated that in a single source the light extinction of the particles is directly related to the particle mass concentration. Scatter in the experimental data can be attributed in part to changes in the particle size distribution. The inability to control the particle size distribution was cited by Engdahl[6] and by Mitchell

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Table I. Light extinction relationships of plumes.

Fractional transmitted light	Ringelmann number	Equivalant opacity Per cent
1.0	0	0
0.8	1	20
0.6	2	40
0.4	3	60
0.2	4	80
0	5	100

and Engdahl[7] as a problem in developing an empirical relationship.

The two general theoretical approaches have been (1) to assume a monodisperse size distribution (or a mean effective particle size) or (2) assume a mathematical relationship for a polydisperse size distribution. Hawksley *et al.*,[8] reported relationships between plume light extinction and mass concentration for an average monodisperse size much smaller than the wavelength of light and for an average monodisperse size much larger than the wavelength of light. Good agreement was reported with the data of Stoecher for the small particle case.[8] Conner and Hodkinson[4] reported the use of a mean effective particle diameter to explain their data and presented calculations relating mass concentration to plume transmittance for average particle sizes in black and white emissions. Robinson[9] reported the use of size distribution data for iron foundry emissions to calculate numerically the effect of control equipment on the plume opacity. The Bay Area Pollution Control Board Regulation 2[10] has a provision limiting the concentration from sources with a relationship developed from a Ringelmann No. 2. Robinson reported that this regulation was developed assuming an oil aerosol with a geometric mass mean radius of 0.23μ and a geometric standard deviation of 3.4. Pilat and Ensor[11] presented general relationships of particulate mass concentration to plume transmittance for log-normal particle size distributions and black and white emissions.

Objectives of This Paper

The objectives of this paper are: (1) to extend the general mathematical relationships between plume opacity and particulate air pollutant properties as reported by Pilat and Ensor to a wider range of variables, (2) to indicate the effect and range of values of particle refractive index and density, and (3) to present recently recorded simultaneous measurements of the plume opacity and particulate properties.

Optical Properties of Particulate Matter

Light Scattering Theory

The attenuation of a collimated beam of light through a turbid medium over a path length L is given by the Bouguer law (Lambert-Beer law)

$$I/I_0 = \exp\{-bL\} \qquad (1)$$

where I/I_0 is the fraction of transmitted light and b is the extinction coefficient of a volume of aerosol (m^{-1}). Equation (1), in a slightly different form, has also been used to relate light transmittance of particle suspensions in liquids. Skinner and Boas-Traube[12] reported an equation in the form

$$I/I_0 = \exp\{-SpWL\} \qquad (2)$$

where Sp is the specific projected particle extinction area (m^2/g) and W is the particulate mass concentration (g/m^3). Skinner and Boas-Traube verified equation (2) for a wide variety of conditions for a transmittance range of about 80–10%. Equations (1) and (2) do not describe the effect of multiple light scattering which may be found in highly concentrated suspensions or over long path lengths. Hodkinson[13] reported that multiple light scattering is rarely important in extinction measurements of aerosols.

The light extinction coefficient b is well defined theoretically and can be used to develop relationships for the specific projected particle extinction area Sp. The extinction coefficient for a volume of aerosol composed of spherical particles is given by

$$b = \pi \int_{r_1}^{r_2} Q_E(\alpha,m) r^2 n(r) dr \qquad (3)$$

where

α = size parameter, $2\pi r/\lambda$
r = particle radius
λ = wavelength of light
m = refractive index of the particles relative to air
$n(r)$ = size frequency distribution, number of particles of radius r per volume per Δr
$Q_E(\alpha,m)$ = particle light extinction efficiency factor, the total light flux scattered and absorbed by a particle divided by the light flux incident on the particle.

The light extinction efficiency factor Q_E for spheres, ellipsoids, and cylinders can be computed using the Mie equations as described by van de Hulst.[15] For pure scatterers with typical refractive indices Q_E can vary from near 0 for very small particles, to about 4 when the particle diameter is near the wavelength of light, and approaches a theoretical limit of 2 for very large particles.

The particulate mass concentration W for spherical particles is given by

$$W = 4/3\pi\rho \int_{r_1}^{r_2} r^3 n(r) dr \qquad (4)$$

where ρ is the average particle density. The particle size frequency distribution $n(r)$ is related to the total particle number concentration N and the particle number fraction frequency $f(r)$ by

$$n(r) = Nf(r) \qquad (5)$$

The fraction of particles between r and $r + dr$ is obtained by multiplying $f(r)$ by the particle radius increment, dr.

Horvath and Charlson[16] reported that the ratio of particulate mass concentration to the extinction coefficient W/b is useful for correlations of atmospheric visibility data. The theoretical W/b ratio from equation (3) and (4) is

$$W/b = \frac{\frac{4}{3}\rho \int_{r_1}^{r_2} r^3 n(r) dr}{\int_{r_1}^{r_2} Q_E(\alpha,m) r^2 n(r) dr} \qquad (6)$$

The ratio can be made concentration independent for single particle scattering by substituting equation (5) into equation (6)

$$W/b = \rho \left[\frac{\frac{4}{3}\int_{r_1}^{r_2} r^3 f(r) dr}{\int_{r_1}^{r_2} Q_E(\alpha,m) r^2 f(r) dr} \right] \qquad (7)$$

For convenience, Pilat and Ensor (1970)[11] assigned the name parameter K to the integral ratio in brackets (specific particulate volume, cm^3/m^3/extinction coefficient m^{-1}). The parameter K is dimensionally similar to the volume surface characteristic size described by Herdan (1960).[16] K is primarily a function of the particle size distribution, refractive index, and to a less degree, the wavelength of light.

Table II. Selected air pollutant size distribution data.

Source, Uncontrolled	Geometric mean radius, r_{gw},μ	Geometric standard deviation, σ_g	Reference
Wood smoke	0.035	1.7	18
Oil fired power plant	0.5	3	19
Electric steel furnace	1.1	8.2	20
Automobile tail pipe	4.6	31	21
Cement dust	8.5	3	22
Pulverized coal power plant	9.5	4	23
Hot mix asphalt	17	2	20
Spreader-stoker coal furnace	35	5	23

From equations (1) and (2), the extinction coefficient b is related to the specific projected area *Sp* and particle mass concentration by

$$b = SpW \quad (8)$$

By substituting equation (7) into equation (8), the specific projected area is given by

$$Sp(m^2/g) = \frac{1}{K(\text{cm}^3/\text{m}^2)\rho(\text{g/cm}^3)} \quad (9)$$

A working equation can be developed from equations (2) and (9) to calculate the expected mass concentration for various values of plume transmittance (or opacity), average particle density, and plume diameter

$$W = -K\frac{\rho}{L}\ln(I/I_0) \quad (10)$$

Range of Important Variables

Particle Size Distribution. The log-normal particle size distribution model can be used to describe a wide variety of polydispersed systems such as those resulting from comminution of solids or spraying of liquids. A complete description of the use of the log-normal size distribution is given by Herdan[16] and by Smith and Jordan.[17] The log-normal size distribution for a number frequency distribution is given by

$$f(r) = \frac{1}{r\sqrt{2\pi}\ln\sigma_g} \times \exp - \left[\frac{\ln^2 r/r_{gn}}{2\ln^2\sigma_g}\right] \quad (11)$$

where r_{gn} is the geometric number mean particle radius and σ_g is the geometric standard deviation (a measure of the polydispersity or breadth of the particle size distribution). Log-normal distributions of particle number, area, and mass are related mathematically. The geometric standard deviation remains the same for these distributions. The relationship between geometric number and mass mean radii, r_{gn} and r_{gw}, respectively is

$$\ln r_{gn} = \ln r_{gw} - 3\ln^2\sigma_g \quad (12)$$

Size distribution data can be reduced graphically by plotting "smaller than" cumulative size frequency versus size on logarithmic probability paper. r_{gw}, for mass distribution data, is the radius at the 50% size and σ_g is given by

$$\sigma_g = \frac{84.13\%\ \text{size}}{50\%\ \text{size}} = \frac{50\%\ \text{size}}{15.87\%\ \text{size}}$$

Particle size distributions reported for air pollutants indicate that both the geometric mass mean radius and the geometric standard deviation can vary over wide ranges. The particle size distribution in the emissions from a given source depends on the nature of the source (combustion, metallurgical, etc.), and the degree of collection. Hopefully, similar sources should have similar particle size distributions. A summary of typical size distributions is presented in Table II. The extremes in the geometric mass mean radius were from 0.04μ for wood smoke to 35μ for fly ash from an uncontrolled coal stoker. The extremes in the geometric standard deviation are 1.0 (monodisperse) to 31 reported for automobile emissions.

Refractive Index. The optical properties of the particles are described by the refractive index. The refractive index is a complex number, $m = n_1 - in_2$. The real part, n_1, describes the light scattering properties and the imaginary part, n_2, describes the light absorption of the particle material. In Table III refractive indices for pure crystalline solids and various liquids are presented. Air pollutant particles may be composed of a mixture of materials.

Particle Density. Densities of pure materials which may be found in air pollutants are also presented in Table III. For actual pollutants the particle density may be much smaller than expected from the pure substance due to unhomogeneous composition of the particle. For example, centospheres (hollow particles) with a density less than 1 g/cc are sometimes found in power plant emissions.[19]

Calculations

The equation for parameter K is obtained by substituting equation (11) for the size frequency distribution $f(r)$ into the integral ratio in equation (7) resulting in

$$K = \frac{4}{3}\frac{\int_{r_1}^{r_2} r^2 \exp - \left[\frac{\ln^2 r/r_{gn}}{2\ln^2\sigma_g}\right]dr}{\int_{r_1}^{r_2} rQ_E(\alpha,m)\exp - \left[\frac{\ln^2 r/r_{gn}}{2\ln^2\sigma_g}\right]dr} \quad (13)$$

The parameter K was calculated using equation (13) with equation (12) to obtain r_{gn} from r_{gw}. The efficiency factor $Q_E(\alpha,m)$ was calculated by forward recursion methods and was in agreement with the results reported by Penndorf.[28] Equation (13) was integrated with the trapezoidal rule, reported by Deirmendjian[30] to be suitable for integrations of light scattering functions over particle size. The value of Δr was about 1% of r over the wide range of particle sizes. The theoretical limit $Q_E = 2.0$ was used for values of α greater than 75. This is a good assumption if the acceptance angle of the instrument used to measure light extinction is very small (for example, the acceptance angle should be less than 0.7° if the largest particle in the duct is 5 μm in diameter[13]).

Results

Calculation of K versus r_{gw} and σ_g

The results for refractive indices of 1.33, 1.50, 1.96–0.66*i*, and 2.8–0.02*i* are presented in Figures 1–4 respectively. These results, as indicated in Table III, cover the range of refractive index which should be important. The refractive index 2.8-0.02*i* is a value assumed to represent a material such as iron oxide with a weak light absorbing component. A wavelength of light of 550 nm was used as an average for visible light. It is approximately the wavelength of maximum sensitivity for the human eye. The range of r_{gw} and σ_g is 0.01 to 100μ and 1.0 (monodisperse) to 10.0, respectively, and should include the values expected for most air pollutants.

The effect of refractive index is relatively unimportant for particle radii greater than about 0.5μ. However, for particle radii less than about 0.5μ, the magnitude of the absorbing index (n_2) is very important. The light extinction by pure light scattering particles (no light absorption $n_2 = 0.0$) decreases rapidly with a decrease in particle size (light scattering is proportional to r^6 for small particles). However, for absorbing or opaque materials, small particles extinguish light proportional to their volume[14] (light extinction is proportional to r^3) resulting in a constant value of K.

Experimental Data

In Table IV experimentally determined optical parameters are presented for various sources. The more optically active the material, the larger is the specific extinction area Sp(m²/g) and the smaller is the specific particulate volume to light extinction coefficient K (cm³/m²). The reporting of both parameters isolates the effect of the particle density assumption. Sp can be computed for a given source with the use of equation (2) and measurements of the light transmittance, particle mass concentration, and path length. K can be computed from Sp with the use of equation (9) and the assuming of an average particle density. K can also be determined with knowledge of the size distribution parameters (geometric mass mean radius r_{gw} and the geometric standard deviation σ_g) and the refractive index from Figures 1–4.

Table III. Physical properties of possible air pollutants.

Substance	Density, g/cc	Temperature, °C	Wavelength of light, nm	Refractive index, $m = n_1 - n_2i$ n_1	n_2	Reference
Aluminum oxide-Al_2O_3	3.5	—	589.3	1.77	10^{-6}	24
Carbon (amorphous)	1.8–2.1	—	436	1.90	0.68	
			546	1.96	0.66	25
			623	2.00	0.66	
Iron oxide Fe_2O_3	5.2	—	589.3	2.78–3.01	?	26
Calcite $CaCO_3$	2.7	—	589.3	1.5–1.65	?	26
Quartz, SiO_2	2.32–2.66	—	589.3	1.544	Very small	26
Na_2SO_4	2.7	—	589.3	1.47	?	26
Zinc oxide ZnO	5.6	—	589.3	2.0	?	26
Wood smoke	1.3	—	540	1.53	0.00095	27
Sulfuric acid 90%	1.811	18.3°	589.3	1.437	Very small	26
Benzene, C_6H_6	0.88	20°	589.3	1.501	Very small	26
n-Decane	0.73	20°	589.3	1.412	Very small	26
Water	1.0	20°	589.3	1.333	Very small	26

Table IV. Measured optical properties of emissions.

Source	$L_{(m)}$	Instrument	Average Sp m²/g	ρ g/cm³ (assumed)	Average K cm³/m²	Reference
Orchard heater (Black smoke)	0.43	Visual	20	2.0	0.025	30
Coal power plant (Fly ash)	1.14	Bolometer	0.78	2.0	0.64	author
Coal stoker (Black smoke)	0.30	Smoke meter	6.1	1.95	0.084	3
Coal stoker (Black smoke)	0.15	Smoke meter	4.6	1.95	0.11	2
Oil power plant (Black smoke)	0.2	Smoke meter	8.7	1.95	0.059	4
White smoke Generator	0.2	Smoke meter		0.87		4
W = 0.22 g/m³			2.5		0.46	
W = 0.47 g/m³			3.8		0.30	
W = 1.00 g/m³			5.75		0.20	
Kraft mill recovery furnace W = 0.15 g/m³	1.52	Bolometer	1.7	1.0	0.6	31
Veneer dryer	1	Visual	2.8	1.0	0.36	32, 33

Discussion

Application of Results

The theoretical approach presented in this paper is intended to be a guide to be used with judgment to estimate plume opacity from the basic physical properties of the aerosol. Obviously, when there are large deviations from the assumptions used in the calculations such as a non-log-normal size distribution model or a change in the aerosol concentration on size distribution between the locations of particle source testing and the plume opacity measurement deviations from the theoretical predictions may result. The measured K data in Table IV indicates the magnitude of the optical activity of actual aerosol emissions and may be used to supplement the theoretical calculation of plume opacity from the particle size distribution. However, the data in Table IV are insufficient to verify the theoretical relationship.

Verification

Each part of the theory should be considered from the standpoint of validity. A fundamental part of our analysis is the use of Mie electromagnetic scattering theory for spherical particles. Mie theory has been verified under very carefully controlled experimental conditions for single spherical particles[34] or clouds of spherical particles.[14] Light extinction by non-spherical particles in random motion was reported by Hodkinson[13] to be nearly the same as that for spherical particles much larger and much smaller than the wavelength of incident light. The effect of irregular particles with sizes near the wavelength of light is to smooth the light extinction efficiency factor Q_E curve as a function of the size parameter α.

Mie theory has been used to calculate the light scattering of polydisperse aerosols for many years.[29] The light scattering coefficient of a cloud of particles was reported by Holland, *et al.*,[35] for talc dust and by Zuev, *et al.*,[36] for artificial fogs to be in good agreement with that calculated with Mie theory using measured particle size distributions and refractive indices.

There remains, however, the verification of the application of Mie theory to the forecasting of plume opacity. Probably the most useful data of all would be that characterizing actual air pollution sources. These data should consist of simultaneous measurements of the light transmittance (or the light extinction coefficient) over a known path length, the particle mass concentration, and the particle size distribution at various efficiencies of the control equipment. It is important to measure the particle size distribution (or characterize it in some way) to indicate if variations in the Ks are due to experimental error or to changes in the particle size distribution. A feature of the theoretical calculations which lends additional credence to the source opacity correlation approach is the relative insensitivity of K to changes in the geometric mass mean particle radius for polydisperse ($\sigma_g > 4.$) size distributions as shown in Figures 1–4. Additionally, the average particle density and the re-

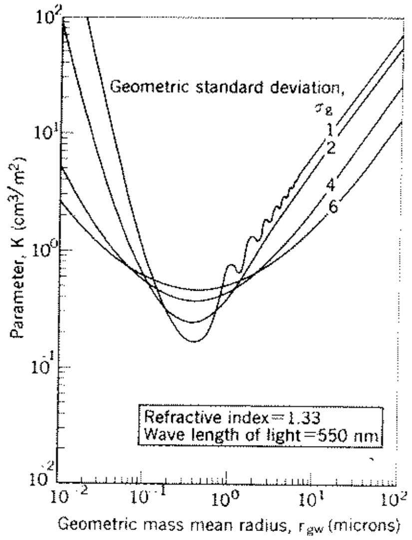

Figure 1. Parameter K as a function of the log-normal size distribution parameters for liquid water.

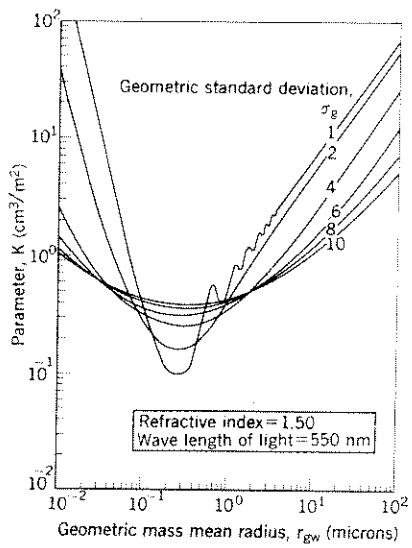

Figure 2. Parameter K as a function of the log-normal size distribution parameters for a white aerosol.

fractive index (the magnitude of the imaginary part is very important for small particles) may not be accurately known for a given source.

Sample Calculation

The estimation of the maximum allowable particulate mass concentration for a given plume opacity standard will be illustrated by an example calculation. Let us assume that the regulation specifies a maximum allowable plume opacity of Ringelmann No. 1 (80% light transmittance through the plume). It is given that the stack diameter L is 32.8 ft (10 m), the exhaust gases are at 300°F, and the particle properties are a mass mean radius r_{gw} of 2μ, a geometric standard deviation σ_g of 3, a density ρ of 2 g/cm³, and a refractive index m of $1.96–0.66i$ (carbon). From Figure 3, a K of 0.6 cm³/m² is obtained. With equation (10) the particle mass concentration which corresponds to the Ringelmann No. 1 is calculated

$$W = \frac{-K\rho \ln (I/I_0)}{L} = \frac{-(0.6 \text{ cm}^3/\text{m}^2)(2 \text{ g/cm}^3) \ln 0.8}{10 \text{ m}}$$

$$W = 0.027 \text{ g/cm}^3 \text{ at stack conditions}$$

Converting to units of gr/ft³ at 300°F

$$W = 0.027 \text{ g/m}^3 \frac{15.43 \text{ gr}}{\text{g}} \times \left(\frac{0.3048\text{m}}{\text{ft}}\right)^3 = 0.012 \text{ gr/ACF}$$

Converting to standard conditions at 60°F

$$W = 0.012 \frac{\text{gr}}{\text{ACF}} \frac{760°\text{R}}{520°\text{R}} = 0.018 \text{ gr/SCF}$$

Therefore for this case the maximum allowable particulate mass concentration which corresponds to a Ringelmann No. 1 is 0.012 gr/ACF or 0.018 gr/SCF. It should be noted that this sample calculation involved an assumption of constant K. Actually there may be some change in K upon the installation of particle collection equipment due to the decrease in the particle size. This variation can be taken into account if information concerning the particle size

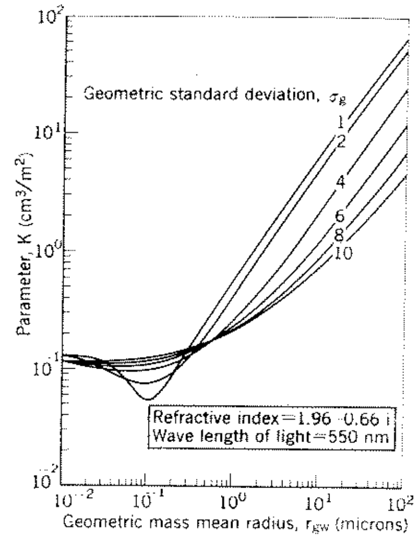

Figure 3. Parameter K as a function of the log-normal size distribution parameters for a black aerosol.

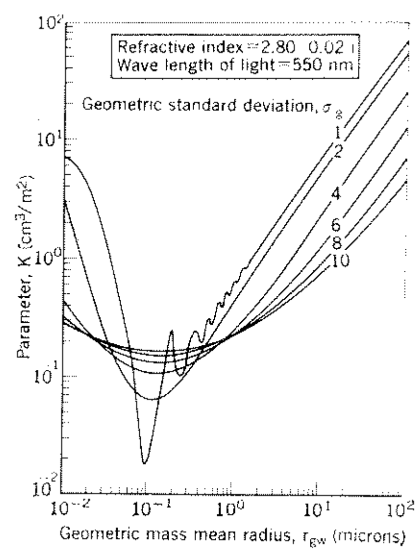

Figure 4. Parameter K as a function of the hypothetical refractive index for iron oxide.

distribution of the controlled emission is available (may be predicted either from particle size measurements at similar sources or by calculations using particle collection efficiencies of control equipment as a function of particle size).

Conclusions

A general theoretical relationship between plume opacity and particle properties has been developed and compared with experimental data. Numerical results have been presented to include the ranges of physical properties expected for air pollutants. The parameter K is primarily a function of particle size for particle radii greater than about 0.5 μm and primarily a function of refractive index for smaller particles. Plume opacity can be estimated for given sources from the numerical results.

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