EUFAR 2nd Summer School on Airborne Cloud and Aerosol Science

Aerosol Optical Properties

by

Andreas Petzold

Institut für Physik der Atmosphäre Deutsches Zentrum für Luft- und Raumfahrt Oberpfaffenhofen 82234 Wessling

andreas.petzold@dlr.de

Aerosol-Climate Interaction

direct effect



Direct effect

resulting from interaction with airborne particles solar radiation is

- (i) scattered back into space
 - \Rightarrow cooling effect,
- (ii) absorbed in the atmosphere

 \Rightarrow heating effect.



Andreas Petzold - ACAS Aerosol Optical Properties

Aerosol-Climate Interaction





$$\Delta \mathbf{R} = \mathbf{R}_{SUM} - \mathbf{R}_{S} \cong \delta \omega_{0} \overline{\beta} \times \{(\mathbf{1} - \mathbf{R}_{S})^{2} - \mathbf{2} \mathbf{R}_{S} \overline{\beta}^{-1} (\omega_{0}^{-1} - \mathbf{1})\}$$

particle optical properties relevant for climate interaction: aerosol optical thickness δ hemispheric backscatter fraction single-scattering albedo

5

Global Aerosol Distribution from Spaceborne Observation



Aerosol Optical Properties are Relevant for ...

Aerosol-Climate Interaction

The impact of aerosol particles on Earth's climate is based on the interaction of particles and hydrometeors (cloud droplets, ice crystals, ...) with visible (solar) and infrared (terrestrial) radiation.

Remote Sensing of Atmospheric Aerosol

The remote sensing of aerosol particles from ground or space by passive (sun photometer, spectro-radiometer) or active (LIDAR) methods is based on the interaction of particles with electromagnetic radiation. Aerosol optical thickness is defined as the integral over the aerosol extinction coefficient.

Size Measurement of Aerosol Particles

The majority of particle sizing methods particularly for the particle diameter size range > 100 nm are based on light scattering methods.



Seinfeld and Pandis Atmospheric Chemistry and Physics, 1998





Seinfeld and Pandis Atmospheric Chemistry and Physics, 1998



© Martin Ebert, TU Darmstadt (mebert@geo.tu-darmstadt.de)

Direct Climate Effect of Anthropogenic Aerosol

Optical Thickness

A Sulfate Aerosols



B Organic Aerosols



C Soil Dust



0. .002.005 .01 .02 .03 .05 .08 .334 -3.89 -.4

Forcing (W/m^2)



Organic Aerosols



Soil Dust



-.1

Surface Temperature (°C) Sulfate Aerosols



Organic Aerosols



Soil Dust

.05 .1 .2 .4 .84.25 -.84 -.2 -.05



.03.05 .1 .2 .4 .8

Hansen et al, Proc. Nat. Acad. Sci. 95, 12753 - 12758.

Remote Sensing of Aerosol Events

MODIS 29 January 2008

Western Sahara Dust Storm Episode 28 - 30 January 2008

Meteosat 2nd Generation 28 January 2008



Meteosat 2nd Generation 28 January 2008



Transport of Atmospheric Aerosol - Dust



Dust layer topped by clouds at 6000 m a.s.l.

MODIS Aerosol Optical Depth 22 January 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INQAA-18 OPRNL DAILY COMP. 1/22/2008 5 - 1/23/2008 5 -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 24 HOURS



MODIS Aerosol Optical Depth 25 January 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INQAA-18 OPRNL DAILY COMP. 1/25/2<u>008_4 - 1/26/2008_4</u> -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 24 HOURS



MODIS Aerosol Optical Depth 28 January 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INCAA-18 OPRNL DAILY COMP. 1/28/2008_4 - 1/29/2008_4 -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 24 HOURS



MODIS Aerosol Optical Depth 29 January 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INCAA-18 OPRNL DAILY COMP. 1/29/2008 4 - 1/30/2008 3 -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 23 HOURS



MODIS Aerosol Optical Depth 31 January 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INQAA-18 OPRNL DAILY COMP. 1/31/2<u>008_3 - 2/1/2008_5</u> -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 26 HOURS



MODIS Aerosol Optical Depth 02 February 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INGAA-18 OPRNL DAILY COMP. 2/2/2008 = 5 - 2/3/2008 = 4 -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 23 HOURS



MODIS Aerosol Optical Depth 04 February 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INGAA-18 OPRNL DAILY COMP. 2/4/2008 4 - 2/5/2008 1 -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 21 HOURS



MODIS Aerosol Optical Depth 05 February 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INQAA-18 OPRNL DAILY COMP. 2/5/2008 1 - 2/6/2008 0 -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 23 HOURS



MODIS Aerosol Optical Depth 06 February 2008

NOAA/NESDIS EDGE IMAGE DISPLAY

OPT. THK. 100KM GLOBAL ANALYSIS INGAA-18 OPRNL DAILY COMP. 2/6/2008 | 0 - 2/7/2008 | 0 -70.0000 TO 70.0000 LAT -180.000 TO 179.000 LON 24 HOURS

Aerosol Optical Properties

The interaction of aerosol particles with electromagnetic radiation gives rise to many spectacular atmospheric effects, such as coloured sunsets, halos around the sun or moon, and rainbows.

The aerosol impact on climate as well as visibility degradation in an aerosolloaded atmosphere is related to the aerosol-radiation interaction.

The predominant fraction of direct-reading real-time aerosol measurement methods uses by some means or other optical detection methods.

The measurement of particle sizes for accumulation mode particles, soil and dust particles as well as for cloud drops and ice crystals is entirely based on optical methods.

Remote sensing methods for aerosols (lidar, satellite-borne radiation spectrometers) rely on the interaction of radiation and particles.

The aerosol-radiation interaction is an efficient method for transferring heat to particles which can then be used for

- particle charging by UV radiation
- particle evaporation for chemical analytical purposes
- measuring combustion particles in flames from incandescence radiation

Andreas Petzold - ACAS Aerosol Optical Properties

Light extinction

The attenuation of light along the direction of propagation through a scattering and absorbing medium is a key process responsible for visibility degradation.

The light attenuation for a parallel beam of light of incident irradiance I_0 is given by the Lambert-Beer law

$$\frac{I}{I_0} = e^{-\sigma_{ext} L}$$

 σ_{ext} = extinction coefficient in m⁻¹; L = length of atmospheric column in m

Mechanisms of interaction

When a beam of light impinges on a particle, electric charges in the particle are excited into oscillatory motion.

The excited electric charges reradiate energy in all directions (*scattering*) and may convert a part of the incident radiation into thermal energy (*absorption*).

Electromagnetic radiation transports energy. The amount of energy crossing an unit area perpendicular to the direction of propagation per time is its irradiance I_0 in W m⁻². The radiation W scattered or absorbed by a particle is proportional to I_0

$$W_{scat} = N C_{scat} I_0$$
 , $W_{abs} = N C_{abs} I_0$

C $_{scat}$ and C $_{abs}$ in units of m² are the single-particle scattering and absorption cross sections.

Conservation of energy requires that the light removed from the incident beam by the particle is accounted for by scattering in all directions and absorption. The combined effect is referred to as *extinction*

$$C_{ext} = C_{scat} + C_{abs}$$

The scattering efficiency of a particle of cross-sectional area A_p is defined as

$$Q_{scat} = C_{scat} / A_{p} \implies Q_{ext} = Q_{scat} + Q_{abs}$$

Andreas Petzold - ACAS Aerosol Optical Properties

The extinction coefficient σ_{ext} for an aerosol consisting of *N* particles per unit volume of cross-sectional area A_p (spherical particle of diameter D_p) is

$$\sigma_{ext} = N A_{p} Q_{ext} = N \frac{\pi D_{p}^{2}}{4} Q_{ext}$$

Particles may extinguish radiation by scattering and/or absorption according to

$$\sigma_{\text{ext}} = \sigma_{\text{scat}} + \sigma_{\text{abs}}$$

The fraction of scattering to extinction is defined by the single-scattering albedo

$$\omega_0 = \frac{\sigma_{scat}}{\sigma_{ext}} = \frac{\sigma_{scat}}{\sigma_{scat} + \sigma_{abs}} = 1 - \frac{\sigma_{abs}}{\sigma_{scat} + \sigma_{abs}}$$

Key Parameters

- ratio of particle size to wavelength of light, or size parameter
- complex index of refraction

for non-absorbing particles is k = 0and for absorbing particles is k > 0*m* is determined by the chemical composition of the particle

$$\alpha = \frac{\pi D_{p}}{\lambda}$$

$$m = n + i k$$

TABLE 22.2 Refractive Indices of Atmospheric Substances at $\lambda = 589$ nm (Unless Otherwise Indicated)

	m = n - ik		
Substance	n	k	<u> </u>
Water	1.333	0 (see Table 22.1)	
Water (ice)	1.309		
NaCl	1.544	0	
H_2SO_4	1.426^{a}	0	
NH4HSO4	1.473 ^b	0	
$(NH_4)_2SO_4$	1.521^{b}	0	
SiO ₂	1.55	0	$(\lambda = 550 \mathrm{nm})$
Carbon	1.96	0.66	$(\lambda = 550 \mathrm{nm})$
Mineral dust ^c	1.56	-0.006	$(\lambda = 550 \mathrm{nm})$

"Stelson (1990), assuming a 97% pure (by mass) mixture of H_2SO_4 with H_2O_2 .

^{*b*}Weast (1987).

'Tegen et al. (1996).

Light-absorbing aerosol
componentsvisible + IRBlack Carbon"green" Fe_2O_3 near IR H_2O
 $(NH_4)_2SO_4$ 9 µmSiO2

In the visible spectral region, only graphitic-like black carbon absorbs light very efficiently;

in the graphite lattice, a twodimensional free electron gas can interact almost perfectly with the electromagnetic radiation.

Scattering and absorption by a sphere

Scattering and absorption cross sections can be calculated exactly for spherical particles by solving the electromagnetic wave equations for a spherical boundary, as presented by Gustav Mie in 1908.

Numerical Mie codes are widely used for the calculation of particle scattering and absorption properties, see Bohren and Huffman, 1983.

Andreas Petzold - ACAS Aerosol Optical Properties

Summary of Mie Theory

(Bohren and Huffmann, 1983)

The angular distribution of scattered light of a polarised plane wave is

$$S_{1} = \sum_{n}^{\infty} \frac{2n+1}{n(n+1)} (a_{n} \pi_{n} + b_{n} \tau_{n})$$
$$S_{2} = \sum_{n}^{\infty} \frac{2n+1}{n(n+1)} (a_{n} \tau_{n} + b_{n} \pi_{n})$$

$$a_{k} = \frac{\alpha \psi_{k}'(\alpha m) \psi_{k}(\alpha) - \alpha m \psi_{k}'(\alpha) \psi_{k}(\alpha m)}{\alpha \psi_{k}'(\alpha m) \varsigma_{k}(\alpha) - \alpha m \varsigma_{k}'(\alpha) \psi_{k}(\alpha m)}$$
$$b_{k} = \frac{\alpha m \psi_{k}'(\alpha m) \psi_{k}(\alpha) - \alpha \psi_{k}'(\alpha) \psi_{k}(\alpha m)}{\alpha m \psi_{k}'(\alpha m) \varsigma_{k}(\alpha) - \alpha \varsigma_{k}'(\alpha) \psi_{k}(\alpha m)}$$

Integral Mie scattering and extinction cross sections are calculated from

$$C_{scat}(m, \alpha) = \int_{0}^{2\pi\pi} \int_{0}^{2\pi\pi} \frac{\left|S(\theta, \phi)\right|^2}{k^2} \sin\theta \, d\theta \, d\phi$$
$$C_{ext}(m, \alpha) = \frac{4\pi}{k^2} \operatorname{Re}\left\{S(\theta = 0^\circ)\right\}$$

$$C_{scat}(m, \alpha) = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \left[\left| a_n \right|^2 + \left| b_n \right|^2 \right]$$
$$C_{ext}(m, \alpha) = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Re} \left\{ a_n + b_n \right\}$$

with wave number *k* and size parameter

$$\alpha = \frac{\pi D_{\rho}}{\lambda}$$

 $\alpha << 1$ Rayleigh scattering regime

 $\alpha \cong 1$ Mie scattering regime

α >> 1 Geometrical optics regime Andreas Petzold - ACAS Aerosol Optical Properties

Rayleigh scattering regime

With respect to visible radiation, particles of diameter $\leq 0.1 \ \mu m$ lie in the Rayleigh regime.

The angular distribution of scattered radiation is symmetrical in the forward and backward direction with respect to the incident light beam and almost independent of particle shape.

Extinction, scattering and absorption efficiencies or small particles

Efficiencies are calculated from the power series expansions of the spherical Bessel functions from Mie theory, when only the first two terms of the power series expansion are considered:

$$Q_{scat}(m,\alpha) = \frac{8}{3} \alpha^4 \left| \frac{m^2 - 1}{m^2 + 2} \right|^2$$
$$Q_{abs}(m,\alpha) \cong 4 \alpha \operatorname{Im} \left\{ \frac{m^2 - 1}{m^2 + 2} \right\}$$

$$Q_{ext}(m, \alpha) = Q_{scat}(m, \alpha) + Q_{abs}(m, \alpha)$$

Thus, for sufficiently small particles

$$Q_{scat}(m,\alpha) \propto \left(\frac{D_{p}}{\lambda}\right)^{4} ; C_{scat} = \frac{\pi D_{p}^{2}}{4} Q_{scat} \Rightarrow \frac{C_{scat}}{V_{p}} \propto \frac{D_{p}^{3}}{\lambda^{4}}$$
$$Q_{abs}(m,\alpha) \propto \frac{D_{p}}{\lambda} ; C_{abs} = \frac{\pi D_{p}^{2}}{4} Q_{abs} \Rightarrow \frac{C_{abs}}{V_{p}} \propto \frac{1}{\lambda}$$

Mass-specific coefficients

Extinction, scattering and absorption efficiencies normalised to particle mass, calculated for green light at $\lambda = 550$ nm:

Rayleigh-regime $D_p \ll \lambda$

 C_{abs} / Volume independent of D_{p} C_{scat} / Volume $\propto D_{p}{}^{3}$

Andreas Petzold - ACAS Aerosol Optical Properties

Measurement of the Extinction Coefficient

The aerosol extinction coefficient is measured in a cavity of sufficient length by determining the light attenuation along the optical path. The light attenuation for a parallel beam of light of incident irradiance I_0 is given by the Lambert-Beer law

$$\frac{I}{I_0} = e^{-\sigma_{ext} L}; C_{ext}(m, \alpha) = \frac{4\pi}{k^2} \operatorname{Re}\left\{S(\theta = 0^\circ)\right\}; \quad \sigma_{ext} = \int C_{ext}(m, \alpha) n_N(D_p) d\log D_p$$

 σ_{ext} = extinction coefficient in m⁻¹; L = length of atmospheric column in m

Long path extinction spectrometer of Research Centre Karlsruhe, Martin Schnaiter.

Measurement of the Scattering Coefficient

The aerosol scattering coefficient is measured in an integrating nephelometer which determines the amount of light scattered in all angles. The light scattering coefficient is given by

$$C_{scat}(m,\alpha) = \int_{0}^{2\pi\pi} \frac{|S(\theta,\phi)|^2}{k^2} \sin\theta \, d\theta \, d\phi \; ; \; \sigma_{scat} = \int C_{scat}(m,\alpha) \, n_N(D_p) \, d\log D_p$$

Integration of scattered light is achieved by collecting light scattered in all solid angles. Data analysis requires correction of truncation angles in the forward direction.

Aerosol Absorption Measurement Methods

Sketches of the major categories of measuring light absorption by aerosol particles, the graph is taken from the Proceedings of the First International Workshop on Light Absorption by Aerosol Particles

(Gerber, H.E. and E.E. Hindman (1982) *Light Absorption by Aerosol Particles*, Spectrum Press, Hampton.).

Filter-based Aerosol Absorption Measurement Methods

- ▷ Sampling of particles on a fibrous filter matrix.
- Measurement of the modification of filter-optical properties by the collected aerosol particles.
- Assumption of Lambert-Beer type relationship for data analysis.

Transmission method

$$\sigma_{0 (REF)} = \frac{1}{2} \frac{A}{V} \ln\left(\frac{R_0}{R}\right)$$

filter surface area A , sampled volume V, mass-specific absorption coefficient $B_{ATN} [m^2 g^{-1}]$, filter mass loading $S_{BC} [\mu g/cm^2]$

Andreas Petzold - ACAS Aerosol Optical Properties

Interaction of Particles, Fibres and Radiation

Filter Matrix Effects

- Multiple scattering of light by filter fibres and light-scattering aerosol particles tends to overestimate the absorption coefficient.
 - "Shadowing" of collected particles inside the fibre matrix tends to underestimate the absorption coefficient.

Multi-Angle Absorption Photometry

The MAAP sensor unit permits the simultaneous analysis of transmittance method, reflectance method and multiangle absorption photometry from the same aerosol sample.

Petzold, A. and M. Schönlinner, J. Aerosol Sci., 35, 421-441, 2004.

RAOS 2002 Kerosene soot experiments

Correction functions for filter loading effects, determined from pure combustion particles; reference absorption coefficient is $\sigma_{ap} = \sigma_{ep} - \sigma_{sp}$.

Correction functions for the effect of aerosol light scattering, determined from kerosene soot ammonium sulphate mixtures

Petzold, A., H. Schloesser, P.J. Sheridan, W.P. Arnott, J.A. Ogren, and A. Virkkula, *Aerosol Sci. Technol.*, 39, 40-51, 2005.

Andreas Petzold - ACAS Aerosol Optical Properties

Particle Sizing by Light Scattering

Rayleigh Regime: Extinction, scattering and absorption efficiencies

Efficiencies are calculated from the power series expansions of the spherical Bessel functions in the coefficients a_n and b_n from Mie theory, when only the first two terms are considered:

$$Q_{scat}(m,\alpha) = \frac{8}{3} \alpha^4 \left| \frac{m^2 - 1}{m^2 + 2} \right|^2$$
$$Q_{abs}(m,\alpha) \cong 4 \alpha \operatorname{Im} \left\{ \frac{m^2 - 1}{m^2 + 2} \right\}$$

Angular distribution of radiation scattered by Rayleigh particles The angular distribution of scattered radiation is symmetrical in the forward and backward direction almost independent of particle shape.

40

Angular distribution of radiation scattered by Mie particles

Polar plots of angular scattering by spheres with m = 1.33 (H₂O).

As size parameter x (α) increases by a factor of 20, forward-tobackward scattering increases by about 1000.

Particle sizing by Mie scattering requires careful modelling of instrument response functions R according to the scattering and detection geometry.

At distinct scattering angles, Mie ambiguities may results in unambiguous size-scattering relationships.

Optical Particle Counters

Principle of operation

- A dilute aerosol flow is directed through a laser beam
- Aerosol particles crossing the laser beam scatter light
- The scattered light is collected in a defined solid angle

Example: The Classical Scattering Aerosol Spectrometer Probe CSASP radiation source He-Ne laser 632 nm: scattering angle 4 - 22°

Instrument	Light source	Light-collecting optics* $\alpha - \beta$	Flow rate or active area
CSASP	5 mW He–Ne laser	4-22	0.15 cm ³ /sec
ASASP	2 mW He-Ne laser (intra-cavity)	4-22	0.1 cm ³ sec
FSSP	5 mW He–Ne laser	3-13	0.25 mm^{2} †
ASSP‡	5 mW He-Ne laser	5.3-12.4°	0.4 mm ² †

Table 2. Characteristics of Knollenberg light scattering aerosol counters

* All instruments have axial symmetry with respect to the direction of the laser source and the polar angles α , β refer to a cone subtending angles α through β from the direction of forward scattering.

† Flow rate can be determined from active area by multiplying by the speed at which air (containing aerosol) passes through the instrument.

[‡] The manufacturer has produced two models of the ASSP having different optics, but only one of these has been studied here. The other collects light scattered $6.7^{\circ}-14.4^{\circ}$ from the direction of forward scattering.

Optical Particle Counters - Calibration Set-Up

The Measurement Process

- 1/ Consider a set of 1000 particles.
- 2/ Determine the size of every particle by appropriate methods.
- 3/ Divide the entire size range into a series of successive particle size intervals
- 4/ Sort the particles according to their size.
- 5/ Plot the grouped data as a histogram of numbers of occurrence.
- ⇒ Simplest form of a number size distribution

Height of the interval = number of particles in the size bin

Width of the interval = upper bound diameter - lower bond diameter

Intervals are not comparable because height depends on width. Andreas Petzold - ACAS Aerosol Optical Properties

Calibration with solid spherical particles of nigrosin dye

Pinnick and Auvermann, J. Aerosol Sci., 10, 55-74, 1979. Andreas Petzold - ACAS Aerosol Optical Properties

Irregularly shaped NaCl particles

Pinnick and Auvermann, J. Aerosol Sci., 10, 55-74, 1979.

Andreas Petzold - ACAS Aerosol Optical Properties

FSSP - 300

Forward Scattering Spectrometer Probe Type 300

scattering angle 5 - 12°

size range D = $0.3 - 20 \,\mu m$

in situ measurement

Probe Characteristics - FSSP

Consider scattering of particles in a plane wave of polarised radiation. Ambiguities of Mie scattering concerning particle size have to be considered.

PCASP 100X

Passive Cavity Aerosol Spectrometer Probe

scattering angle 35 - 120°

size range D = 0.1 - 3 μ m

measures dry aerosol

active sampling of aerosol

focusing of the aerosol by a dry sheath air flow

intensive radiation field generated by multiple reflections of the laser beam in a passive cavity outside the laser resonator.

Andreas Petzold - ACAS Aerosol Optical Properties

Probe Characteristics - PCASP

Consider scattering of particles in a wave inside a reflector. Since mutiple reflection in the passive cavity do not generate coherent radiation, no standing wave is generated.

INPUT DATA

Application #2: Optical Closure for Dust

Saharan dust layer over Ouarzazate (Morocco) during SAMUM-1.

Application #2: Optical Closure for Dust

Saharan dust samples over Ouarzazate (Morocco) during SAMUM-1.

Application #2: MISR validation on 19 May 2006

Application #3: Aerosol Type Identification

Raileigh regime
$$Q_{ap}(m, \alpha) \propto \frac{D_p}{\lambda} \operatorname{Im} \left\{ \frac{m^2 - 1}{m^2 + 2} \right\}$$

if the complex refractive index is almost independent of $\lambda,$ as is the case for soot, then

if the complex refractive index shows a strong $\lambda\text{-depence},$ as for dust, then

å_{ap}≅1

å_{ap} > 3

#080128ab to Tenerife

MODIS 29 January 2008

Western Sahara Dust Storm Episode 28 - 30 January

#080129ab Dakar plume

5 100 90 80 4 70 Absorption Angström 1/Mm 3 60 PSAP raw , my man man man 50 40 2 30 20 1 10 0 0 14.5 14.7 14.9 15.1 15.3 15.5 15.7 **Dec Time** Teneriffe changing type of dust to Praia

Application #3: Aerosol Type Identification

Application #3: Aerosol Type Identification