The mechanisms of aerosols and radiation interactions (Aerosol Optics)

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> Aerosol Radiation interaction (aerosol optics)

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Aerosol Optics

Interaction of light with particles.

Light is electromagentic wave particles contain atoms---> positive and negative charges electric field of wave exerts force on charges.

A neutral atom consists of positive nucleus and negative electrons. Under influence of electric field center of positive and negative charges is displaced, a dipole is formed.

Dipole changes its dipole moment with frequency of light wave. Oscillating dipole emits light:

Light is **scattered** by the particle, i.e. part of the incident light is deflected in other directions.

Other possibility for interaction of light with particle. **absorption** Vibration of dipoles is damped, Part of the light energy is converted to internal energy of the particle. Aerosol Radiation interaction (aerosol optics)

Light scattering



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Light scattering





elastic scattering

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Part or all of the incident light is "lost", i.e. Converted to other forms of energy (e.g. Heat)

For particles scattering and absorption occurs simultaneaously

Interaction of light with particles is characterized by three magnitudes: Scattering coefficient σ_s absorption cofficient σ_a Volume scattering function $\gamma(\Theta)$ (additional information needed for polarization) and magnitudes derived from these: Phase function P(Θ) Single scattering Albedo ω Absorption number a Definitions will be given in the following

The exponential attenuation of light is well known. Usually written as $\Phi = \Phi_0 \exp(-k.x)$

 Φ, Φ_0 incident parallel light flux and emerging parallel light flux

- x Distance, the flux passes through the medium
- k absorptive properties for medium (unit m^{-1})

very similar for aerosol:

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Incident light

Aerosol

transmitted light

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 $\Phi = \Phi o \exp (-\sigma_e.x)$

 σ_e extinction coefficient Unit: m⁻¹, km⁻¹, Mm⁻¹

 Φ/Φ_0 is called the transmission T, therefore T = exp($-\sigma_e.x$) Aerosol Radiation interaction (aerosol optics) Light attenuation can take place in two ways: Scattering absorption Therefore the extinction coefficient σ_e is the sum of the scattering coefficient σ_s and the absorption coefficient $\sigma_{a:}$

 $\sigma_e = \sigma_s + \sigma_a$

 σ_s can be understood as the part of attenuation due to scattering and σ_a can be understood as the part of attenuation due to absorption

BUT: Scattering and absorption always take place simultaneously. Only if the material of the particles is completely transparent (e.g. NaCl in the visible) only scattering occurs. The light scattered usually is different in different directions. The angular dependence is characterized by the volume scattering function.

But before this we have to introduce the radiometric units (based on energy). Photometric units based on human sensation, will not be used here

Radiant flux Φ:

Energy of radiation per second passing through a (real or imaginative) surface. Flux Φ , unit $[\Phi] = W$ (1W is the unit of power)

Example 1: Flux emitted by a light bulb. Imaginative surface e.g. sphere around bulb

Example 2: Light flux entering the window of a photocell: Surface is entrance window of cell



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Radiant flux density S: Flux transiting a unit surface: $S=d\Phi/dA$, unit $[S] = W/m^2$

Radiation is considered incident from all directions in a hemisphere (or emitted) onto a surface that coincides with the base of that hemisphere.

Other names: Irradiance (E) Radiant emittance

Example 1: Total radiation (direct and diffuse, visible, UV and IR) reaching 1 m² of ground. e.g. 1000 W/m², (depends of time of day, time of year, clouds, geographical latitude, etc.)

Example 2: Radiation emitted by black body of temperature T: $S = \sigma . T^4$ (Stefan-Boltzmann law).

If the radiant flux is a parallel beam of light:

dA is assumed to be perpendicular to direction of flux.

Example: Solar Constant: radiant energy passing per second through a surface of 1 m² perpendicular to the direction of radiation:

extraterrestrial (top of atmosphere): $S = 1360 \text{ W/m}^2$

In general radiative sources do not emit radiation equally in all directions:

We consider an angular range where homogeneous emission takes place i.e. a small solid angle.

Solid angle is defined in the following way:



Consider a sphere of radius R around source.

The angular range $d\omega$ intersects the sphere (any shape possible), the surface area is dA

Solid angle is dA/R^2 . Unit: Steradian sr, (no dimension)

(aerosol optics)

Full solid angle: $4.\pi R^2/R^2 = 4.\pi$ Half space, e.g. above a a plane: $2.\pi$. **Radiant Intensity** is defined as light flux per solid angle $I = d\Phi/d\omega$, Unit: W/sr

Attention: Intensity is used as a (wrong) synonym for many other magnitudes !!

Radiant intensity works well for point (or nearly point) sources. For extended sources we use the **radiance**.



Summary:

QUANTITY	RADIOMETRIC	
power	watt (W)	Flux
power per unit area	W/m^2	Flux density
power per unit solid angle	W/sr	Radiative intensity
power per unit area per unit solid angle	W/m^2 -sr	Radiance

Now back to the scattering of light by particles: Consider a volume element of particles



Light scattered by the particles is removed from the incident beam,

therefore:

$$\sigma_s = \int_{4\pi} \gamma(\theta) d\omega$$

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Can be shown easily: Consider dV=dx.dA,

S=d\Phi/dA and
$$d\Phi = -\Phi \cdot \int_{4\pi} \gamma(\theta) d\omega \cdot dx$$

For rotational symmetry:
$$d\omega = 2\pi sin\theta d\theta \longrightarrow \sigma_s = 2\pi \int_0^{\pi} \gamma \sin\theta d\theta$$

Now: how to obtain $\sigma_e, \sigma_s, \sigma_a, \gamma(\theta)$ theoretically?

Treat the interaction of light with a material containing dipoles. We already know: the dipoles in the particle change the dipole moment due to the vibration of the electric vector of the light wave. Simple case: Particle is much smaller than the wavelength of light $(<\lambda/20)$:

---> all dipoles vibrate in phase, Radiation simply is the sum of the radiation of all dipoles. First treated by Lord Rayleigh

Result: Scattering coefficient:
$$\sigma_s = N \cdot \frac{128\pi^5 r^6}{3\lambda^4} \left(\frac{m^2 - 1}{m^2 + 1}\right)^2$$

N number of particles r radius of particle λ wavelength of light m refractive index of particle material

Aerosol Radiation intera20on (aerosol optics) Volume scattering function:

$$\gamma(\theta) = N \cdot \frac{8\pi^4 r^6}{\lambda^4} \left(\frac{m^2 - 1}{m^2 + 1}\right)^2 \cdot [1 + \cos^2(\theta)]$$

Similar formulas for gas molecules (also $<<<\lambda$)

Discussion:

(a) Scattering proportional to r⁶,
 ---->the smaller the particle, the less scattering (half size, 1/64 scattering)

(b) Scattering proportional to λ⁻⁴,
 ----> light with shorter wavelength is scattered stronger (blue light scattered more than red, e.g. blue sky or blue oils smoke)

(c) light is partly polarized: The scattering function of the particle has two parts: (1) the scattered light polarized perpendicular to the scattering plane or plane of observation (*plane formed by the direction of incident light and scattered light*) Aerosol Rad



This part is: $N \cdot \frac{8\pi^4 r^6}{\lambda^4} \left(\frac{m^2 - 1}{m^2 + 1}\right)^2 \cdot [1]$

 $m \dots$ refractive index

Scattered light flux independent of scattering angle θ .



No scattering at 90°

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Raleigh scattering is completely polarized (perpendicular) at 90^{23}

For particles $>\lambda/20$ The dipoles cannot be considered as oszyllating in phase. Therefore constructive and destructive interference occurs, depending on size, refractive index, wavelength of light and direction. The radiation emerging from the particle is more complicated and other ways of treating the scattering and absorption of light are needed.

For spherical particles a rigorous solution has been found by G. Mie (1908) by solving the Maxwell equations. (For non spherical particles see below).

Particles are characterized by their size r and the refractive index m.

Optical properties of the bulk material are characterized by -- refractive index n (for Snell's law)

-- bulk absorption coefficient α (transmission of bulk material of length x is exp($-\alpha$.x)

conductors (metals, graphite) are characterized by a complex refractive index

 $m = n - i \cdot k$ (m = n + i \cdot k in Bohren and Huffman)

The imaginary part - k characterizes the the absorptive properties of the material.

Relation to α :

$$k = \alpha \cdot \lambda / (4 \pi)$$

Aerosol Radiation interaction (aerosol optics) Example : Black glass is 2 mm thick and has transmission 0.001 (considered strongly absorbing)

Bulk absorption coefficient is $\alpha = 3454 \text{ m}^{-1}$.

complex refractive index is $m = 1.5 - i \cdot 0.000154$

For comparison: Complex refractive index of graphite is

 $m = 2.0 - i \cdot 1.0$ (one of many values !!!!, see below)

Strong absorbers in bulk need not to have a large imaginary part.

Light attenuation by particles suspended in a gas.

First: simple geometric optics:

Consider an area of 1 m² containing *n* black circles of radius r and Flux Φ incident



The flux intercepted by the black circles is given by the ratio of the covered surface and the total surface.



Re-interpretation of the previous: consider a thin slab of aerosol of thickness dx



containing N particles $/ m^3$

Number of Particles in slab having a cross section of 1m² is n = N. Volume = N . 1 m^2 . dx

Extinction coefficient σ_{e}

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Similar as before:

 $\sigma_{e} = N \cdot r^{2} \pi$ $d\Phi = -\Phi . N . 1m^2 . dx r^2 \pi / 1 m^2 = -\Phi . N . r^2 \pi dx$ Aerosol Radiation interaction (aerosol optics)

Valid for particles much larger than wavelength:

. . . .

For small particles: Diffusion on the edges, scattering inside particles, interference between wave going through and around particle,

Therfore $\sigma_{e}{=}~N~$. $r^{2}~\pi$. Q_{e}

 Q_e is called the extinction (scattering, absorption) efficiency factor. It tells how much more (less) of the light incident on the particle cross section is attenuated / scattered / absorbed

Q factor depends on ratio of wavelength to particle size given by size parameter $x = 2 r \pi / \lambda$ (size parameter also called α) and the refractive index m of the material of the particle

Many computer programs to calculate Qe, Qs, Qa are availabe, e.g. Bohren C.F., Huffman D.R. (1998) Absorption and Scattering of Light by Small Particles. Wiley Interscience, 544 pp., , Barber P.W., Hill S.C. (1990) Light Scattering by Particles: Computational Methods World scientific, 261pp or 29 http://diogenes.iwt.uni-bremen.de/vt/laser/wriedt/index_ns.html). Aerosol Radiation interaction

Example:

extinction, scattering, and absorption efficiency



Why oszyllations?

Wave around particle : distance 2r is $2r/\lambda$ wavelengths Wave through particle: Wavelenght in medium is λ/n therfore n.2r/ λ wavelengths

Difference in number of wavelengths: $2r (n-1)/\lambda$ (reduced size parameter)

If this is $\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$: destuctive interference. Since particle is transparent: more light is scattered in other direction: constructive interference in these directions.

Our example n=1.5, $2r/\lambda$ (n-1)=1/2 or $2r\pi/\lambda$ (n-1)=1/2. π Or 2r π/λ = x = π =3.14 maximum attenuation Other values: $x = \pi = 3.14$, $x = 3\pi = 9.5$, $x = 5\pi = 15.7$

Actually for cube, for sphere slightly larger x: 4.5, 11.5, 17.5

Remarks: (a) for real m and small x: the Q_s factor goes to zero proportional to x^4 Rayleigh scatter σ_s proportional to λ^{-4} and r^6

(b) For strongly absorbing material and small x: the Q_a factor is proportinal to x

Why?? (Moosmueller et al. JQSRT 2009)

(a) Scattering: particle divided in many small elements. Each element becomes a dipole due to electric field. Since particle is very small, no phase difference, the electric field by all dipoles is added \rightarrow electric field of all dipoles is proportion to volume, i.e. $\vec{E} \propto r^3$ Light flux proportinal to $E^2 \propto r^6$ Since $Q_s = (\text{scattered flux}) / (\text{cross section}) \rightarrow Q_s \propto r^4$ since $x=2r\pi/\lambda \rightarrow Q_s \propto x^4$. Scale invariance:

scattering depends on ratio of size and wavelength Larger particle and larger wavelength: same conditions. $\rightarrow Q_s$ must depend on x only, since $Q_s \propto x^{-4}$ and $x=2r\pi/\lambda$ \rightarrow thus $Q_s \propto \lambda^{-4}$ (refractive index constant)

(b) Absorption:

is damping of the oszyllation of the dipoles. i.e. transfer of the energy of the electromagnetic wave into heat.

This is an incoherent process.

Each element absorbs a certain amount of light flux.

Since number of elements proportional to volume: absorption coefficient propertinal to r^3 , therefore $Q_a \propto r^1 \propto x^1$.

extinction, scattering, and absorption efficiency



Qe, Qs, Qa

Example:

extinction, scattering, and absorption efficiency



for m = 1.5 (most substances) and size parameter = 3 (particles with diameter of 0.5 μ m, normally main fraction in atmospheric aerosol) the Q factor increases (linear) with x

----> more scatter in blue, less in red

-----> red sunset

for large x the Q factor approaches 2, particles attenuate twice the quantity of the light incident on the particle (i¿ i¿ i¿ paradox!?!?!?) Solution: consider also light diffracted on the edges Absorbing particles have no oszyllations, (resonances are damped).

For an aerosol consisting of particles of different sizes, characterized by a number size distribution the extinction, scattering and absorption coefficient is obtained by integration:

$$\sigma_{e,s,a} = \int \pi r^2 Q_{e,s,a}(x,m) n(r) dr$$

Since the Q factor depends on the size parameter ($x = 2r\pi/\lambda$) which includes the wavelength, the coefficients depend on wavelength.

Frequently the extinction coefficient decreases with increasing wavelength

In many cases an empirical relation for $\sigma_e(\lambda)$ holds:

$$\sigma_e(\lambda) = \beta \cdot \left(\frac{\lambda}{\lambda_0}\right)^{-\alpha}$$
Ångström Formula,
 $\lambda_0 \dots$ reference wavelength, usually 1µm
 α , [sometimes (- α)] Ångström Exponent

Light absorption coefficient can be obtained by using Q_a . If particles are light absorbing, the scattered light usually is less. For strongly absorbing particles the scattering and the absorption both amount to about half of the extinction.

In the atmosphere we have many particles originating from different sources and both extinction coefficient, scattering coefficient and absorption coefficient are the sum of the respective coefficients of the different species, i.e. $\sigma_{e,s,a} = \sum_{i=1}^{n} (\sigma_{e,s,a})_i$

with $(\sigma_{e,s,a})_i$ the extinction, scattering, absorption coefficient of species i. This type of mixing of particles is called an external mixture.

But also each particles can also consist of a mixture of various substances (internal mixing). This frequently happens if particles agglomerate due to diffusion/coagulation, or are incorporated in cloud droplets, where other substances are adsorbed 40

Particles fromed e.g. during conbustion an a diesel engine

(1)coagulate to agglomerates

(2)combustion gases cool and organic vapors condense, also further oxidation

800 800 000





-- (4)Further condensation



- -- (5)Incorporation in cloud droplet
- -- (6)Chemical reactions on the surface of the droplet
- -- (7)etc.
- -- (8)re-evaporation

MIXED PARTICLE

A theoretical treatment of the optical properties of such particles are difficult to impossible (see end of lecture)

Frequently spherical particles with an effective (or average) refractive index are used. This is the refractive index a homogeneous spherical particle would have in order to have the same optical properties as the considered internally mixed particle.

One of the mixing rules is the volume mixing rule:

$$\rho = \Sigma \rho_i \cdot v_i$$
, $m = \Sigma n_i \cdot v_i - i \cdot \Sigma k_i \cdot v_i$. v_i volume fraction

Many other rules exist

CAUTION: Effective medium, just like other rules, has no theoretical basis (but still gives insight in the problem! Chylek et al, 1988 44

Aerosol Radiation interact

Both the extinction coefficient, scattering coefficient and the absorption coefficient increase with particle concentration. It is convenient to know which fraction of the intercepted light which is scattered and absorbed respectively. This is given by dimensionless numbers which are independent of the number of particles:

Single scattering albedo $\omega = \sigma_s / \sigma_e$ Absorption number: $a = \sigma_a / \sigma_e$ obviously $a + \omega = 1$

Example: Hazy day in Vienna: $\sigma_e = 0.39 \text{ km}^{-1}$, $\sigma_a = 50 \text{ Mm}^{-1}$

 $\sigma_s = 390 \text{ Mm}^{-1} - 50 \text{ Mm}^{-1} = 340 \text{ Mm}^{-1}$, therefore $\omega = 340 / 390 = 0.872$, a = 50 / 390 = 0.128

Angular scattering:

Mie theory (computer programs) give the volume scattering function. For N spherical particles per m³ of radius r for illumination with non polarized light:

 $\gamma(\theta) = N \ \lambda^2 / (4 \ \pi) \ . \frac{1}{2} (i_1 + i_2),$

 i_1, i_2 intensity function for scattering of light with polarization perpendicular and parallel to the scattering plane. Both are functions of size parameter x, refractive index m and scattering angle θ



For larger particles ($r = 0.8 \mu m$ at $\lambda = 550 nm$) complicated structure of scattering. Only visible for strictly monodisperse particles. For micrometer sizes (r = 0.26 μm at $\lambda = 550 nm$) forward scattering strongly

increases.

For small sizes ($r = 0.03 \ \mu m$ at $\lambda = 0.55 \ \mu m$) the scattering is similar to Rayleigh Scattering: i_1 almost constant, i has minimum at 90°

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 i_2 has minimum at 90°

Aerosol Radiation interaction (aerosol optics) Volume scattering function proportional to particle number per m³.

If only the angular dependence is of interest: use normalized scattering function:

4
$$\pi$$
. $\frac{\gamma(\theta)}{\int \gamma(\theta) d\omega} = P(\theta)$

... Phase function, normalized scattering function

Phase function is used with and without the factor 4π (Attention!!), we will use it with 4π .

Example: phase functions for non polarized light

Phase function for spherical particles with refractive index 1.5 - 0.0 i and size parameters 0.3, 3, and 9



 $\gamma(\theta)$, P(θ) usually depend on wavelength, i.e. $\gamma(\theta, \lambda)$, P(θ, λ)

Since
$$P(\theta) = 4\pi \cdot \frac{\gamma(\theta)}{\int_{4\pi} \gamma(\theta) d\omega}$$

 σ_s
therefore $\gamma(\theta) = \frac{1}{\sigma} \cdot P(\theta)$

therefore
$$\gamma(\theta) = \frac{1}{4\pi}\sigma_s \cdot P(\theta)$$

since $\sigma_s = \omega \cdot \sigma_e$

$$\gamma(\theta) = \frac{1}{4\pi} \cdot \omega \cdot \sigma_e \cdot P(\theta)$$

Aerosol Radiation interaction 49 (aerosol optics) Depending on the size of the particles the scattering of the light is distributed inhomogeneously in the different directions. A completely isotropic medium would have $P(\theta) = 1$.

The symmetry or non-symmetry of the scattering can be described by the asymmetry parameter g, defined as:

$$g = \frac{\int \gamma(\theta) \cos(\theta) d\omega}{\int \gamma(\theta) d\omega} = \frac{\int_0^{2\pi} \gamma(\theta) \cos(\theta) \sin(\theta) d\theta}{\int_0^{2\pi} \gamma(\theta) \sin(\theta) d\theta}$$

The asymmetry parameter is zero for completely symmetric scattering, one for pure forward scattering and -1 for pure back scattering. For the phase functions on the previous slide, the asymmetry parameters are: 0.0701, 0.734, and 0.648, respectively

Asymmetry parameter is diffcult to measure (full phase function needed)

Backscatterd fraction can be measured with an intergrating nephelometer:

$$b = \frac{\int_{\pi/2}^{\pi} \gamma(\theta) \sin(\theta) d\theta}{\int_{0}^{\pi} \gamma(\theta) \sin(\theta) d\theta}$$

Obvious: less backscatter, more asymmetric scattering,

m = 1.5 - 0.01 i



i.e. b \downarrow then g \uparrow

m = 1.5 -0.01 i



Comparison phase function spheres - spheroids

Phase function spheres vs. spheroids (1:2)





Non-spherical non-homogeneous particles.

What to do with mixed non spherical particles???



- -- not spherical
- -- not homogeneous
- -- each particle looks different
- -- random orientation



Example for atmospheric particle originating in the Gobi desert and transported 2000 km to Japan. Takes up sulfate, black carbon, nitrates, ... Aerosol Radiation interaction 55 (aerosol optics) Spherical particles with effective medium approximation is just a first guess (still may be helpful)

For simple shapes (spheroids, cylinders, bispheres, Chebyshev particles) exact solutions exist. (e.g. Mishchenko et al, 2002)

With powerful computers one can do better:

E.g. Discrete Dipole Approximation DDA:

Particle of arbitrary shape: Volume of particle is digitized. Volume is divided into regions sufficiently small that the interaction with the electromagnetic wave in this region can be represented by one single dipole of dipole moment given by the material (refractive index). Interaction of all dipoles (also with each other) can be calculated. Needs a super computer, but is possible.



Other example:

Irregular particles of this shape,

having mass size distribution





Gives the following phase function:

Experimental data confirm the shape of the DDA curve, but do not agree perfectly:



From: Angular scattering of the Gobi Desert aerosol and its influence on radiative forcing by H. Horvath; M. Kasahara; S. Tohno; M. Kocifaj *Journal of Aerosol Science* **37**, **#10** pp. 1287-1302, (2006). 59

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Figure 7 of H. Horvath, L. Alados Arboledas, F. J. Olmo Reyes(2018) Angular Scattering of the Sahara Dust Aerosol *Atmos. Chem. Phys.* **8**, 17735-17744, 2018 https://doi.org/10.5194/acp-18-17735-2018 Aerosol Radiation interaction 61 (aerosol optics)





Attenuation law: $\Phi = \Phi_0 \cdot e^{-\sigma_e \cdot x}$ Single scattering albedo $\omega = \frac{\sigma_s}{\sigma_s}$ Extinction coefficient scattering coefficient absorption coefficient abs

$$\sigma_e = \int_{r_{min}}^{r_{max}} r^2 \pi . n(r) . Q_e(m, x) . dr \qquad x = \frac{2r\pi}{\lambda} \quad \text{(size parameter)}$$

Volume scattering function
$$\gamma(\theta) = \frac{d\Phi}{dV.d\omega}$$
, $\sigma_s = \int_{4\pi} \gamma(\theta) d\omega$

Phase function
$$P(\theta) = \frac{1}{4\pi} \cdot \frac{\gamma(\theta)}{\sigma_s}$$
 Asymmetry parameter $g = \frac{\int_0^{\pi} \gamma(\theta) \cdot \sin(\theta) \cdot \cos(\theta) \cdot d\theta}{\int_0^{\pi} \gamma(\theta) \cdot \sin(\theta) \cdot d\theta}$
Aerosol Radiation interaction $b = \frac{\int_{\pi/2}^{\pi} \gamma(\theta) \sin(\theta) \, d\theta}{\int_0^{\pi} \gamma(\theta) \sin(\theta) \, d\theta}$ 63