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Aerosol Basics: Definitions, size distributions, structure

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Aerosol particles

- Liquid or solid particles suspended in a carrier gas
- Described by their
 - Size
 - Concentration
 - Number
 - Surface
 - Mass
 - Volume
 - Chemical composition
- Lifetime seconds years (in troposphere typically hours days)

The Earth's energy balance



Kiehl and Trenberth, 1997



Climate effects of aerosols

Direct effects

- Scattering and absorbing short- and longwave radiation
- Examples: sulphate, organic carbon, black carbon, aerosols from biomass burning, mineral dust
- Indirect effects
 - Cloud formation: effectiveness of the aerosol acting as cloud condensation nuclei (CCN)
 - Depend on: size, chemical composition, ambient environment
- Big uncertainty in the effects
 - Longest continuous measurement time series 12 years

RADIATIVE FORCING COMPONENTS





Aerosols, clouds and climate







more CCNless precipitationchange in cloudmore scatteringhigher albedodynamics



Visibility



Antarctica: > 100 km

New Delhi: < 1.5 km

Pictures: Ismo K. Koponen and Petteri Mönkkönen



Atmospheric Brown Cloud (Pollution)





MODIS 3.5.2006 09:29 http://www.sat. dundee.ac.uk/



Effect on air quality (Helsinki, August 2006)



Photos:

Pia Anttila, FMI



Loss in life expectancy attributable to exposure to fine particulate matter - 2000



Current estimates show that aerosol particles have significant effect on both life expectancy and life quality

Loss in statistical life expectancy that can be attributed to the identified anthropogenic contribution to PM2.5 (months), for the emissions of the year 2000. Calculation results for the meteorological conditions of 1997. Source: IIASA



Types of Aerosols

- Based on their <u>formation processes</u>, aerosols are either <u>primary</u> or <u>secondary</u>:
 - Primary aerosols are directly emitted to the atmosphere.
 - Secondary aerosols are formed in the atmosphere by gas-to-particle conversion processes:
- Based on their <u>sources</u>, aerosols are either natural or anthropogenic:
 - Natural aerosols are emitted as a result of processes in the nature (windblown dust, pollen, plant fragments, seasalt, seaspray, volcanic emissions)
 - Anthropogenic aerosols are somehow related to human activities (fossil fuel burning, industrial processes, traffic, burning of biomass or biofuel, agricultural activities, etc.)



Figure by Mikko Moisio and Ilona Riipinen



Structures





Asbestos



In model calculations, the shape of an aerosol particle is assumed spherical

- In practice, this is not always the case
- There are several ways of characterizing real particles with a certain diameter so that some of their features correspond to the features of a spherical particle of the given size
 - Electrical mobility
 - Terminal velocity
 - Density



Coal





Volcanoes





Variation of Aerosol Concentrations

■ Concentration varies depending on location and time → high concentrations are encountered when there are nearby sources.

	PN [cm ⁻³]	$PM_{10} [\mu g/m^3]$		
Lower troposphere:				
-Urban traffic	> 100 000	> 20		
-Urban background	10 000 - 50 000	20 - 100		
-Rural	500 - 10 000	10 - 50		
-Marine	200 - 5000	5-20		
-Remote	50 - 500	0.02 – 1		
Free troposphere	100 - 100 000			
Stratosphere (background)	1 – 20			



Aerosol size distributions

- The total concentration of atmospheric aerosol particles can vary over 7 orders of magnitude (~10¹ – ~10⁸ #/cm³)
- The size range spans over 5 orders of magnitude (~1 nm – ~100 μm)
- Size affects both the lifetime and the physical and chemical properties
- How to describe the aerosol size and number/area/volume in a simple way? → Aerosol size distribution



Figure by Hanna Vehkamäki and Veli-Matti Kerminen



Lognormal distribution function

It has been observed that atmospheric aerosols can be described rather well with a set of log-normal distribution functions (log-normal = normally distributed in logarithmic scale)

$$n_N \left(\log D_p \right) = \sum_{i=1}^n \frac{N_i}{\left(2\pi \right)^{1/2} \log \sigma_i} \exp \left(-\frac{\left(\log D_p - \log \overline{D}_{pi} \right)^2}{2 \log^2 \sigma_i} \right)$$

 $n_N(\log D_p)$: number of particles of diameter D_p

 \overline{D}_{pi} : median diameter of the mode

 N_i : number concentration of particles in the mode

 σ_i : geometric standard deviation



Representations of Aerosol Concentrations

- Aerosol particle
 concentrations can be
 expressed by Number,
 Surface area, Volume, or
 Mass per unit volume:
- The number concentration is (in most cases) dominated by the ultrafine aerosols.
- The mass or volume concentration is dominated by the coarse and accumulation aerosols.



Representation of number and volume aerosol size distributions Figure from Seinfeld & Pandis, 2006



- Mixture of primary emissions from industry, transportation, power generation, and natural sources and secondary aerosols through gas-to-particle conversion
- Number concentration dominated by ultrafine particles
- Surface area mostly in the 0.1-0.5 µm sizes
- Mass typically has two dominating modes: accumulation and coarse
- Huge variation depending on the measurement site and current meteorological conditions





- Mainly of natural origin, but with some influence of anthropogenic sources
- Number concentration typically has two dominating modes in the ultrafine size range
- Surface area mostly in the 0.1-0.5 µm sizes
- Mass dominated by coarse mode





Remote continental aerosol

- Mainly natural primary particles including dust, pollen, plant waxes and secondary oxidation products
- Number concentration typically has two dominating modes (nucleation mode, accumulation mode)
- Surface area mostly in accumulation mode
- Mass dominated by accumulation mode



distribution. Figure from Seinfeld & Pandis, 2006



Marine aerosol

- Mostly of marine origin: evaporation of seaspray, seasalt, secondary aerosols formed after oxidation of dimethyl sulfide emitted by phytoplankton
- Number concentration typically has two dominating modes around 60 nm and 200 nm
- Mass dominated by coarse mode





Free tropospheric aerosol

- Mid- and upper troposphere above clouds
- Modes around 10 nm and 250 nm
- Number concentration of accumulation mode particles typically higher than in the lower troposphere
 - No precipitation scavenging
- Nucleation mode often present
 - Suitable conditions for new particle formation



Typical free tropospheric aerosol size distribution. Figure from Seinfeld & Pandis, 2006



- Very low total concentrations
 Accumulation mode dominates
 "Arctic haze" during the winter and early spring: anthropogenic sources
- Composition: aged carbonaceous aerosols originated from midlatitude pollution sources, sulfate, seasalt, mineral dust



Typical polar aerosol size distribution. Figure from Seinfeld & Pandis, 2006



- Three overlapping modes at 10 nm, 50 nm, and 10 µm
- Surface area and volume strongly dominated by windblown sand
- Individual dust storms can transfer desert aerosol over the ocean



Typical desert aerosol size distribution. Figure from Seinfeld & Pandis, 2006



Parameters for model aerosol distributions

TABLE 0.0 Turan		Mode I			Mode II		Mode III		
Type	$\frac{N}{(\text{cm}^{-3})}$	<i>D_p</i> (μm)	log σ	$\frac{N}{(\text{cm}^{-3})}$	<i>D_p</i> (μm)	$\log \sigma$	<i>N</i> (cm ⁻³)	<i>D_p</i> (μm)	$\log \sigma$
Urban Marine Rural	9.93×10^4 133 6650	0.013 0.008 0.015	0.245 0.657 0.225	1.11×10^{3} 66.6 147	0.014 0.266 0.054	0.666 0.210 0.557	3.64×10^4 3.1 1990	0.05 0.58 0.084	0.337 0.396 0.266 0.380
Remote continental Free troposphere Polar	3200 129 21.7 726	0.02 0.007 0.138 0.002	0.161 0.645 0.245 0.247	2900 59.7 0.186 114	0.116 0.250 0.75 0.038	0.217 0.253 0.300 0.770	0.3 63.5 3×10^{-4} 0.178	0.52 8.6 21.6	0.380 0.425 0.291 0.438

TABLE 8.3 Parameters for Model Aerosol Distributions Expressed as the Sum of Three Lognormal Modes

Source: Jaenicke (1993).



Processes Modifying Atmospheric Aerosols

Processes affecting the concentration and other properties (size, chemical composition) of atmospheric aerosols include:

- Emissions (primary particles, emissions of aerosol precursor gases)
- Atmospheric transportation
- Deposition from the atmosphere to surfaces (ground, vegetation, water)
- Aerosol dynamics and chemistry



- RH > 100%
 - Due to cooling (isobaric/adiabatic)
- Isobaric cooling (pressure remains constant)
 - Radiative losses of energy, horizontal movement of an airmass over a colder land surface or colder airmass
- Adiabatic cooling (no heat exchange)
 Ascending air parcel pressure decrease, volume expansion, temperature decrease



Cloud/fog formation

- Droplet formation without existing nuclei would require considerable supersaturations
 - e.g. pure water: RH 300-500%
- Aerosol particles that can facilitate droplet formation at low supersaturations are called cloud condensation nuclei (CCN)

$$N_{activated} = f(C, T, RH, w, dist)$$

$$N_{a$$



Köhler theory

Two effects

- A smaller droplet has higher equilibrium vapour pressure due to curvature (Kelvin effect)
- A solution has lower equilibrium vapour pressure than a pure substance
- Köhler theory: combination of two expressions
 - Kelvin equation
 - Raoult's law

$$\ln\left(\frac{p_w(D_p)}{p^\circ}\right) = \frac{A_w}{D_p} - \frac{B_s}{D_p^3}$$

$$A_w = \frac{4M_w\sigma_w}{RT\rho_w} \qquad B_s = \frac{6\nu_s n_s M_w}{\pi\rho_w}$$

Modified versions of the Köhler theory available for soluble trace gases, slightly soluble substances and surface-active solutes



Köhler curves





Cloud droplet composition

Liquids

- Water (solvent)
- Dissolved compounds, e.g. O₃(aq), H₂O₂(aq)
- lons, e.g. SO₄²⁻, NH₄⁺
- Solids
 - Soluble compounds, e.g. ammonium sulfate
 - Slightly soluble compounds, e.g. organic acids, calcium sulfate
 - Insoluble compounds, e.g. dust, elemental carbon
- Condensing gases
 - e.g. H₂O, HNO₃, NH₃, SO₂, H₂O₂



Mixing state of the aerosol

- Important when determining e.g. CCN solubility or aerosol optical properties
- External mixture: each particle from only one source
- Internal mixture: all the particles of a certain size contain a uniform mixture of components from each source

An example of time evolution of size distribution in Hyytiälä





A new particle formation event

 New particles appearing in the 3-25 nm size range (A)

 Newborn particles growing, sometimes to sizes where they can act as cloud condensation nuclei (B)



Figure by Miikka Dal Maso and Ilona Riipinen



Condensation sink (CS) $CS = 4\pi D \int_{0}^{\infty} r \beta_{M}(r) n(r) dr = 4\pi D \sum_{i} \beta_{M} r_{i} N_{i}$

describes the aerosol population's ability to remove vapor by condensation

Coagulation sink (CoagS(D_p))
I describes the aerosol population's ability to remove particles
of size D_p
CoagS(D_p) = $\int_{D_p}^{\infty} K(D_p, D_p', T, ...) n(D_p) dD_p$

Sinks sensitive to particle size changes

hygroscopic growth must be accounted for