# Optical properties of atmospheric constituents

**Direct effects of aerosols optical properties on climate** 





Clouds evaporation

Graphics: Yinon Rudich





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# Climatic effects of aerosols: Large uncertainties



# Optically important Aeroslos in the atmosphere

- Air molecules
- Black carbon aerosols
- Soil dust aerosols
- Organic aerosols
- Aerosols of marine origin
- Sulfates Nitrates Sulfates Nitrates SCATTERERS Organics Soil ABSORBER Elemental Carbon

• H<sub>2</sub>O

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Different approaches to study aerosols OP:

1. Lab (isolated, possibly modified)

2.Inversion of in-situ measurements (no control, hard to isolate specific contributions)

Aerosols science is extremely complex due to wide variety of different types and aging effects. Scattering and absorption crosssections of aerosols:



Bohren and Clothaux, 2006.

**Figure 2.12:** Absorption cross sections for most of the strongly absorbing atmospheric gases (1013 mb and 294 K) and average scattering cross section for air molecules (top panel).

Absorption lines are broadened by: Doppler (motion of molecules and source of photons)  $\infty T$ 

Self broadening (consequence of intermolecular forces) ∝T

Foreign broadening (interactions with other molecules) ∝pressure, T

→ Expect absorption
spectra to vary with
Pressure and Temperature in
the atmosphere.

**10**<sup>-13</sup> Absorption Cross Section  $(\mu m^2)$ **10**<sup>-14</sup> 1013 mb **10**<sup>-15</sup> 10<sup>-16</sup> 10<sup>-17</sup> 10 mb **10**<sup>-18</sup> **10**<sup>-19</sup> 10<sup>-20</sup> 10-21 1085.7 1085.5 1085.3 1084.9 1085.1 1084.7 Wavenumber (cm<sup>-1</sup>)

**Figure 2.19:** Line-by-line calculations of water vapor absorption at two pressure altitudes. The dashed curve shows a calculation from which continuum absorption is omitted. At 10 mb, omission of the continuum results in a curve indistinguishable from that shown.

Bohren and Clothaux, 2006.

Note: wavenumber=1/wavelength=v/c

Effect of size of drops on scattering properties of cloud:



From Kopeika, 2002

Example: Mie output for a black carbon aerosol (sensitivity to size for the same composition)



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# Light scattering experiment



From presentation by Volten



Volten H, Muñoz O, Rol E, de Haan JF, Vassen W, Hovenier JW, Muinonen K, Nousiainen T. Journal of Geophysical Research, 106, 17375-17401,2001

From presentation by Volten

# Absorption by dust particles when suspended in water using an integrating sphere:

| ID               | Description                        | Origin                                                                  | SPM (g m <sup>-3</sup> ) | POC (g m <sup>-3</sup> ) |
|------------------|------------------------------------|-------------------------------------------------------------------------|--------------------------|--------------------------|
| ILL <sub>1</sub> | Illite                             | Source Clay Minerals Repository, University of<br>Missouri (ref. IMt-1) | 37.71                    |                          |
| ILL <sub>2</sub> | As above but different<br>PSD      | As above                                                                | 26.53                    | 0.86                     |
| KAO1             | Kaolinite (poorly<br>crystallized) | As above, but ref. KGa-2                                                | 28.34                    |                          |
| KAO <sub>2</sub> | As above but different<br>PSD      | As above                                                                | 23.3                     |                          |
| MON <sub>1</sub> | Ca-montmorillonite                 | As above, but ref. SAz-1                                                | 38.48                    |                          |
| MON <sub>2</sub> | As above but different<br>PSD      | As above                                                                | 28.52                    | 0.73                     |
| CAL <sub>1</sub> | Calcite                            | Natural crystal                                                         | 26.48                    |                          |
| CAL <sub>2</sub> | As above but different<br>PSD      | As above                                                                | 12.60                    |                          |
| QUA <sub>1</sub> | Quartz                             | Natural crystal                                                         | 14.17                    |                          |
| SAH1             | Atmospheric dust from<br>Sahara    | Red rain event, Villefranche-sur-Mer, France, Nov<br>1996               | 43.43                    | 1.28                     |
| SAH <sub>2</sub> | As above but different<br>PSD      | As above                                                                | 19.47                    |                          |
| AUS <sub>1</sub> | Surface soil dust                  | Cliff shore, Palm Beach north of Sydney, Australia                      | 32.03                    | 0.87                     |
| AUS <sub>2</sub> | As above but different<br>PSD      | As above                                                                | 24.05                    | 0.64                     |
| ICE1             | Ice-rafted particles               | Glacier runoff, Kongsfjord, Spitsbergen, Norway                         | 28.51                    | 0.44                     |
| ICE2             | As above but different<br>PSD      | As above                                                                | 22.37                    |                          |
| OAH <sub>1</sub> | Surface soil dust                  | Oahu, Hawaii Islands                                                    | 31.67                    | 1.12                     |
| OAH <sub>2</sub> | As above but different<br>PSD      | As above                                                                | 18.05                    | 0.62                     |
| KUW <sub>1</sub> | Surface soil dust                  | Kuwait (eastern part, close to ocean)                                   | 23.36                    |                          |
| KUW <sub>2</sub> | As above but different<br>PSD      | As above                                                                | 23.10                    | 6.27                     |
| NIG <sub>1</sub> | Surface soil dust                  | Southwest Nigeria                                                       | 37.06                    | 2.71                     |
| SAN <sub>1</sub> | Atmospheric dust                   | San Diego, California                                                   | 17.08                    | 2.73                     |

### Why did they suspend them in water?



Fig. 3. (a, b) Spectra of mass-specific absorption coefficient of particles for samples of mixed particulate assemblages and (c) for samples of single-mineral species.

Stramski et al. 2007



## <u>Complex</u> refractive index of single composition aerosol

Scattering only

### Strongly absorbing aerosol



Retrieval by Mie (A≈d) calculation – no *a-priori* assumption of refractive index From presentation by Y. Rudich

## Angstrom exponent of HULIS samples: 3 to 5



High spectral dependence. Hoffer et. al. measured Å=7 (BB HULIS), Kirchstetter measured Å=2.5 (OC extracts)

From presentation by Y. Rudich

#### Shape effects on the phase function:

)8

DUBOVIK ET AL.: DUST REMOTE SENSING USING SPHEROID MODEL



Figure 3. (left)  $P_{11}(\Theta, \lambda, \varepsilon_p)$ , the phase functions of desert dust simulated for the spheroid model with 11 single axis ratios  $\varepsilon_p$ . (right)  $P_{11}(\Theta, \lambda)$ , the phase function of desert dust simulated as a mixture of spheroids with different  $\varepsilon_p$  (blue line).  $P_{11}(\Theta, \lambda)$  is obtained by integrations of  $P_{11}(\Theta, \lambda, \varepsilon_p)$  with axis ratio distribution  $dn(\varepsilon_p)/d\ln\varepsilon$ . Also shown is  $P_{11}(\Theta, \lambda)$ , the phase function of desert dust simulated under assumption of spherical particles (red line). All phase functions were simulated with the size distribution and complex refractive index of the Saudi Arabia desert dust model [*Dubovik et al.*, 2002a].



**Figure 8.** Results of fitting a Feldspar scattering matrix measured at 0.441  $\mu$ m. The blue line shows the simulation based on the assumption of spheroidal particle shape. The red line shows scattering matrices simulated for spherical particles using the size distribution and complex refractive index retrieved using the spheroid model.

## Soot Dus Ammonium Sulfate **Sulfuric Acid Marine Organic** Sea Salt **Biomass Smoke** 1µm Pollen // Nucleation Aitken Accumulation Coarse Volume Number

### More on aerosols: variation in size , shape and composition:

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1000

1000

100

Particle Diameter, nm

10

1

### Mass (hence size) distribution of atmospheric particles:



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How relative humidity change optical properties of soluble substances – soluble organic material and sea



At high relative humidity , organic soluble material have higher volume and more material is dissolved, therefore its properties change.

# Continental Common Aerosols

Olga Sima and Ran Vardimon (taken from "OPAC")

Continental and urban aerosol mixtures consist of 3 main aerosol types: Water soluble (~60%) - sulfates, nitrates and other organic substances insoluble (~40%) - mostly soil particles with a certain amount of organic material soot (<1%) - well, soot... (absorbing black carbon)

Insoluble Soluble Soot 2,500 1.8E+7 35,000 crmalized Coe... [cm^2/nicrogram] 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1'20, 1' 1.6E+7 Normalized Coefficient Coefficient Normalized Coefficient 30,000 microgram] cm^2/microgram] 1.4E+7 25,000 1.2E+7 20,000 1.0E+7 Vormalized 8.0E+6 15,000 5 6.0E+6 10,000 4.0E+6 È 5,000 2.0E+6 0 0 0.0E+0 scattering scattering 500 1000 2000 1500 scattering 1500 500 1000 2000 0 1000 2000 absorption absorption absorption Axis Title wavelength [nm] wavelength [nm] - attenuation attenuation -attenuation Rmod =  $0.03 \,\mu m$ Rmod =  $0.5 \,\mu m$ 

We compared the attenuation coefficients (normalized per concentration):

#### **Insoluble particles attenuation is dominant!**

All three aerosol types have lognormal size distributions. Soot (black) has higher absorption coefficient (> scattering). Rmod =  $0.01 \,\mu m$ 



Size Distribution

Method: Mie modeling based on microphysical data.

#### Inverse approaches:

Sunlight changes as it is going through the atmosphere. Observing the sun directly at specific wavelength provides for atmospheric integrated optical depth  $\rightarrow$  concentration (e.g. water vapor). Calculations from remote sensing involve two atmospheric passages.

No vertical distribution information.



AERONET (AErosol RObotic NETwork):



### AOD:

Sun photometer measurements of the direct (collimated) solar radiation provide information to calculate the columnar aerosol optical depth (AOD). AOD can be used to compute columnar water vapor (Precipitable Water) and estimate the aerosol size using the Angstrom parameter relationship.

### ATMOSPHERIC TRANSMISSION





Plot from\_GEOG-1425 class notes, Katherine Kink, University of Minnesota. Note inverted x-scale. Almucantar (circle on the celestial sphere parallel to the horizon) measurements:

Measurements of cloud free day angular distribution of sky radiance + AOD + RT calculations are used to obtain:

Particulate size distribution Index of refraction (real and imaginary) Spectral single scattering albedo

Requires consideration of three main components:

- 1. Gaseous absorption (avoided by choice of  $\lambda$ , and use of climatologies).
- 2. Molecular scattering (calculated for given Pressure).
- 3. Aerosol absorption and scattering.

Minor (ignored) components: ground albedo, stratification



Figure 8: Cimel performing a GOSUN procedure

Use libraries of single particles optical properties (Mie or other)

Needs: RT model Optimum inversion scheme

## **LEVEL 2 Retrievals**



## Level-2 Non-Spherical Aerosol Checks and Processing





Summary of aerosol optical properties retrieved from worldwide AERONET network of ground-based radiometers.

# Biomass-Burning Aerosols\*

- Biomass-burning in Zambia (savanna) during the dry season (Aug.-Sep.) of 1997
- Measurement done with AERONET
- SSA ( $\omega_0$ ) = 0.82–0.85 (at 550nm)

# Size Distribution



Figure 5. Aerosol volume size distributions for biomass burning aerosols in Mongu, Zambia, for August 25 to September 7, 1997, for aerosol optical depth at 440 nm varying from 0.25 to 1.53. Aerosol size distribution retrievals were derived from simultaneous analysis of sky radiances in the almucantar and spectral  $\tau_a$  at 440, 675, 870, and 1020 nm.

# Single Scattering Albedo



Figure 10. Comparison of spectral (440 to 1020 nm)  $\omega_0$  retrievals made with AERONET radiometers at three sites, for  $\tau_a$  at 440 nm ranging from 0.66 to 0.89. The sites are Mongu, Zambia, with savanna burning smoke, Concepcion, Bolivia, with pasture/forest burning smoke in Amazonia, and Goddard Space Flight Center, Maryland, with urban/industrial pollution. See Figure 6 for a comparison of the aerosol volume size distribution for these same sites.





From Kopeika, 2002

# Conclusions

- 1. Atmospheric OP are complex
- 2. Creative approaches provide possibility to retrieve AOPs from routine observations.
- 3. Need for validation (laboratory work is critical).
- 4. Much work is left for the next generation to tease out (e.g. 'anomalous' absorption, absorption by clouds).