







Observing aerosols Federico Fierli Contribution from many colleagues and co-workers

Acknowledge the teaching material from D. Jacob – U-Harvard – S. Massie NCAR





"We have in this fine dust [aerosols] a most beautiful illustration of how the little things in the world work great effects by virtue of their numbers."

John Aitken (1839-1919)

-John Aitken, 1880

Why should we care about aerosol and dust ?

Human health

Air quality

Global Climate Radiation, Chemistry, Rainfall

Aerosols are important from the molecular to the global scale

Aerosols and human health



1952: the "London smog disaster"

Air pollution O₃, CO, NO₂, SO₂ aerosol



Aerosols are the principle component of what we perceive as "smog"

Submicron aerosols are primarily responsible for visibility reduction.





Pasadena, CA, on a clear day (hills are 7 km away

Environmental Protection Agency (EPA) $PM_{2.5}$ 15 µg /m³ (annual average) PM_{10} 150 µg /m³ (24 hour) regulations.

See national map of compliance at: http://en.wikipedia.org/wiki/File:Pm25-24asuper.gif

Pasadena, CA, on a bad smog day

Aerosols and human health

Submicron aerosols can penetrate to the deepest parts of the lung whereupon they can affect the pulmonary part of the respiratory system. For this reason many, including the EPA, consider them dangerous air pollutants.



The role of atmospheric aerosol on meningitis spread





Source	Estimated Flux (Tg yr ⁻¹)			2
	Low	High	Best	Particle Size Category ^a
NATURAL				
Primary				
Soil dust (mineral aerosol)	1000	3000	1500	Mainly coarse
Sea salt	1000	10000	1300	Coarse
Volcanic dust	4	10000	30	Coarse
Biological debris	26	80	50	Coarse
Secondary				
Sulfates from biogenic gases	80	150	130	Fine
Sulfates from volcanic SO ₂	5	60	20	Fine
Organic matter from biogenic VOC	40	200	60	Fine
Nitrates from NO_x	15	50	30	Fine and coarse
Total natural	2200	23500	3100	
ANTHROPOGENIC				
Primary				
Industrial dust, etc. (except soot)	40	130	100	Fine and coarse
Soot	5	20	10	Mainly fine
Secondary				
Sulfates from SO ₂	170	250	190	Fine
Biomass burning	60	150	90	Fine
Nitrates from NO_x	25	65	50	Mainly coarse
Organics from anthropogenic VOC	5	25	10	Fine
Total anthropogenic	300	650	450	
Total	2500	24000	3600	n na cheann. A' fhair ann an ann ann an Solainn a' seann an s

TABLE 2.19 Global Emission Estimates for Major Aerosol Types in the 1980s

"Coarse and fine size categories refer to mean particle diameter above and below 1 $\mu\text{m},$ respectively.

Note: Sulfates and nitrates are assumed to occur as ammonium salts. Flux unit: Tg yr⁻¹ (dry mass).

Source: Kiehl and Rodhe (1995).

Seinfeld and Pandis

450 / 3100

= 15%

Annual mean PM_{2.5} concentration (2002) derived from MODIS satellite instrument data





Aerosols : solid and liquid particles suspended in the air

Size: nm to 100 microns (range of 10⁵) Lifetime: Troposphere (days to weeks) Stratosphere (year)

Primary aerosol: emitted directly into the air Secondary aerosol: gas to particle conversion

Composition: sulfate, ammonium, nitrate, sodium, trace metals, carbonaceous, crustal, water

Carbonaceous

elemental: emitted directly into the air (e.g. diesel soot) organic: a) directly by sources (e.g combustion, plant leaf) b) condensation of low volatile organic gases "fine" diameters D < 2.5 microns
sulfate, ammonium, organic carbon, elemental carbon
Nuclei mode 0.005 to 0.01 microns
condensation of vapors
Accumulation mode
0.1 to 2.5 microns

"coarse" diameters D > 2.5 microns natural dust (e.g. desert) mechanical processes crustal materials biogenic (pollen, plant fragmets)

Aerosol Types and Origin



- Aerosol particles larger than about 1 μm in size are produced by windblown dust and sea salt from sea spray and bursting bubbles.
- Aerosols smaller than 1 µm are mostly formed by condensation processes such as conversion of sulfur dioxide (SO₂) gas (released from volcanic eruptions) to sulfate particles and by formation of soot and smoke during burning processes.
- After formation, the aerosols are mixed and transported by atmospheric motions and are primarily removed by cloud and

precipitation processes.

Aerosol Size Distribution

It presents 3 modes :

 - « nucleation »: radius is between 0.002 and 0.05 mm.
They result from combustion processes, photo-chemical reactions, etc.

 - « accumulation »: radius is between 0.05 mm and 0.5 mm. Coagulation processes.

« coarse »: larger than 1 mm.
From mechanical processes like aeolian erosion.

« fine » particles (nucleation and accumulation) result from anthropogenic activities, coarse particles come from natural processes.



Size Distribution

Log-normal particle size distribution

dN/dr = Σ (N_i / (2π) ^{1/2} r lnσ) exp {- (ln r/ln r₀)²/2ln²σ)

Units: number per cm⁻³ per microns

i=1,2 2 modes

N_i, total number of particles for mode i (cm⁻³)

- r, radii (microns)
- σ , mode width (dimensionless)
- r₀, modal radii (microns)

Aerosols come from a variety of sources, and reside in the atmosphere for weeks



Seinfeld and Pandis



Seinfeld and Pandis

Key aerosol microphysical parameters

Particle size and size distribution

Aerosol particles > 1 mm in size are produced by windblown dust and sea salt from sea spray and bursting bubbles. Aerosols smaller than 1 mm are mostly formed by condensation processes such as conversion of sulfur dioxide (SO₂) gas to sulfate particles and by formation of soot and smoke during burning processes

Effective radius

Moment of size distribution weighted by particle area and number density distribution

Complex refractive index

The real part mainly affects scattering and the imaginary part mainly affects absorption

Particle shape

Aerosol particles can be liquid or solid, and therefore spherical or nonspherical. The most common nonspherical particles are dust and cirrus The bimodal nature of the size-number distribution of atmospheric particles suggests at least two distinct mechanisms of formation, and the chemical composition of the particles reflects their origins.

Fine particles have a diameter smaller than about 2.5 mm, and are produced by the condensation of vapors, accumulation, and coagulation. They have a chemical composition that reflects the condensable trace gases in the atmosphere: SO₂, NH₃, HNO₃, VOC's, and H₂O. The chemical composition is water with SO₄⁻², NO₃⁻, NH₄⁺, Pb, Cl⁻, Br⁻, C(soot), and organic matter; where biomass burning is prevalent, K⁺.

Coarse Particles have a diameter greater than about 2.5 mm, are produced by mechanical weathering of surface materials. Their lifetimes, controlled by fallout and washout, are generally short. The composition of particles in this size range reflects that of the earth's surface - silicate (SiO₂), iron and aluminum oxides, CaCO₃ and MgCO₃, over the oceans , NaCl.

Global dust emissions (modeled)



- A natural laboratory to study
- (i) the contribution of long-range transport,
- (ii) the ageing of continental air masses over the basin,
- (iii) the impact of aerosol on the regional climate,
- (iv) the impact of atmos. deposition on low-Chl, low-nutrient surface waters



Dust event – how it works ?



Figure 1. Up (a,b) True color images from MODIS-Terra (left) and MODIS-Aqua (right) on 22 Ma 2018. The arrows indicate the location of Crete; Down (c,d) MSG-SEVIRI dust RGB images on 1200 U (left) and 1500 UTC (right), 22 March 2018.

Figure 5. (a) Frontal activity, temperature (°C) and wind vectors at 925 hPa and (b) Near-surface dust concentration (μ g m⁻³) and wind vectors at 10 m (zoom from the external 12 × 12 km domain), on 22 March 2018 0600 UTC.

(a)

(b)

Dust event – how frequent is it ?





Fig. 4. (a) Top: mean frequency of African dust outbreaks (%) across the Mediterranean Basin during the period 2001–2011; (b) bottom: mean frequency of African dust outbreaks (%) versus latitude across the Mediterranean Basin during the period 2001–2011.

Dust event – impacts on Particulate Matter

4.100

1402

J. Pey et al.: African dust outbreaks over the Mediterranean Basin during



Fig. 7. Seasonal partitioning of PM_{10} (in $\mu g m^{-3}$), considering the influence of African dust (average values for the periods v available, in most cases from 2001–2010) across the Mediterranean Basin. NAF: African dust outbreaks.

J. Pey et al.: African dust outbreaks over the Mediterranea

J. Pey et al.: African dust



Fig. 2. Annual PM_{10} levels (µg m⁻³) at regional (RB) and suburban (SUB) background sites across the Mediterranean for the period 2001–2011.



Fig. Al. Annual PM_{10} levels ($\mu g m^{-3}$), without African dust, at regional (RB) and suburban (SUB) background sites across the Mediterranean for the period 2001–2011. NAF: African dust outbreaks.

Aerosols and climate



← Drives Global Warming

Direct effect – Light is scattered and absorbed

IPCC, AR4

Table 1. Overview of the different aerosol indirect effects.³

Effect	C loud Type	Description	Sign of the Radiative Forcing
First indirect aerosol effect (cloud albedo or Twomey effect)	All clouds	For the same cloud water or ice content more but smaller cloud particles reflect more solar radiation	Negative
Second indirect aerosol effect (cloud lifetime or Albrecht effect)	Warm clouds	Smaller cloud droplets decrease the precipitation efficiency thereby prolonging cloud lifetime	Negative
Semi-direct effect	Warm clouds	Absorption of solar radiation by soot leads to an evaporation of cloud droplets	Positive
Glaciation indirect effect	Mixed-phase clouds	An increase in ice nuclei increases the precipitation efficiency	Positive
Thermodynamic effect	Mixed-phase clouds	Smaller cloud droplets inhibit freezing causing supercooled droplets to extend to colder temperatures	Unknown
Surface energy budget effect	All clouds	The aerosol induced increase in cloud optical thickness decreases the amount of solar radiation reaching the surface, changing the surface energy budget	Negative

"Warm clouds" clouds with liquid droplets "Mixed phase clouds" clouds with liquid and ice

Aerosol-radiation interactions

Scattering aerosols



Aerosols scatter solar radiation. Less solar radiation reaches the surface, which leads to a localised cooling.



The atmospheric circulation and mixing processes spread the cooling regionally and in the vertical.

Absorbing aerosols



Aerosols absorb solar radiation. This heats the aerosol layer but the surface, which receives less solar radiation, can cool locally.



At the larger scale there is a net warming of the surface and atmosphere because the atmospheric circulation and mixing processes redistribute the thermal energy.

Aerosol-cloud interactions

Aerosol-cloud interactions



Aerosols serve as cloud condensation nuclei upon which liquid droplets can form. More aerosols result in a larger concentration of smaller droplets, leading to a brighter cloud. However there are many other possible aerosol-cloud-precipitation processes which may amplify or dampen this effect.

IPCC AR5, Chapter 7, 2013

Aerosol radiative forcings



IPCC AR5, Chapter 7, 2013

Aerosol "indirect effect" on climate





clean cloud (few particles): large cloud droplets

- low albedo
- efficient precipitation

polluted cloud (many particles): small cloud droplets

- high albedo
- suppressed precipitation (very controversial)

Aerosols and climate: indirect effect

"Ship Tracks" off the coast of Washington



- aerosols are the "seeds" upon which water vapor condenses to form a cloud (these are called "cloud condensation nuclei, or CCN).
- If people make more aerosols, we make more cloud droplets, but because there is a fixed amount of water vapor in the air these droplets will be smaller.
- smaller droplets scatter light more efficiently!
- smaller cloud droplets may also impact rain from these clouds.
- very difficult effect to observe and model!

Observational needs

If your concern is ...

Mass transported through the air for biogeochemical cycles, then you want to know the mean diameter of the particles with the mass or volume. In other words, "What size particles carry the most mass?"

loss of visibility then you want to know the diameter of the particles that have the largest cross section or surface area. In other words, "What size particles cover the largest surface area?"

Cloud formation or microphysics then you want to know the range of diameters with the largest number of particles. In other words, "What is the size of the most abundant particles?"

Observational needs

If your concern is...

human health then you need to know about both the mass and number of the particles, because only a certain size particle can enter the lungs.

Direct climate effect then you need to know about global distribution and impact on radiative budget

Observational needs



There is no perfect instrument – they observe a specific aspect at a different time and spatial scale ...

Ground-based – reference stations



Gravimetric method – reference for air quality monitoring Advantage: direct measure of mass – reliable Disadvantage: relatively expensive, need for operator, not global coverage

https://www.eionet.europa.eu/etcs/etc-atni/products/etc-atni-reports/etcacm_tp_2011_21_pm10equivalence

Ground-based – optical instruments



https://doi.org/10.4209/aaqr.2017.12.0611
Remote Sensing Overview

- What is "remote sensing"?
 - Using artificial devices, rather than our eyes, to observe or measure things from a distance without disturbing the intervening medium
 - It enables us to observe & measure things on spatial, spectral, & temporal scales that otherwise would not be possible
 - It allows us to observe our environment using a consistent set of measurements throughout the globe, without prejudice associated with national boundaries and accuracy of datasets or timeliness of reporting
- How is remote sensing done?
 - Electromagnetic spectrum
 - Passive sensors from the ultraviolet to the microwave
 - Active sensors such as radars and lidars
 - Satellite, airborne, and surface sensors
 - Training and validation sites

The Electromagnetic Spectrum



- Remote sensing uses the radiant energy that is reflected and emitted from Earth at various "wavelengths" of the electromagnetic spectrum
- Our eyes are only sensitive to the "visible light" portion of the EM spectrum
- Why do we use nonvisible wavelengths?

Key aerosol optical parameters

Optical depth negative logarithm of the direct-beam transmittance column integrated measure of the amount of extinction (absorption + scattering)

Single-scattering albedo v_0 given an interaction between a photon and a particle, the probability

that the photon is scattered in some direction, rather than absorbed

Scattering phase function

probability per unit solid angle that a photon is scattered into a particular direction relative to the direction of the incident beam

Angstrom exponent a exponent of power law representation of extinction vs. wavelength



Optical Depth

 $I = I_0 \exp \left[-(\sigma_{\text{scattering}} + \sigma_{\text{absorption}}) (n L) \right] \quad \text{Beer's Law}$

- I_0 original intensity of light that goes into the cell
- I observed intensity of light after it travels through the cell
- L, path length of cell (cm)
- n, number density of particles (cm⁻³)

 $\begin{aligned} & \tau = (\sigma_{\text{scattering}} + \sigma_{\text{absorption}}) \text{ (n L) } & \text{Optical depth (unitless)} \\ & a = (\sigma_{\text{scattering}}) / (\sigma_{\text{scattering}} + \sigma_{\text{absorption}}) & \text{Single scattering albedo} \\ & \text{K} = (\sigma_{\text{scattering}} + \sigma_{\text{absorption}}) \text{ n } & \text{Extinction coefficient (1 / km)} \end{aligned}$

Optical Properties

 $\beta_{\text{ext}}(\lambda) = 10^3 \int Q_{\text{ext}}(r,\lambda) \pi r^2 \, dN/dr \, dr$ extinction (units km⁻¹)

r particle radius (μ m) dn / dr particle size distribution (# cm⁻³ μ m⁻¹)

 $\textbf{Q}_{\text{ext}}\left(\textbf{r}\text{,}\lambda\right)~\text{extinction efficiency (from Mie theory)}$

 $Q_{ext} = Q_{sca} + Q_{abs}$ (sca=scattering, abs = absorption)

Q is a function of the complex index of refraction i.e. composition

Size parameter x = 2 π r / λ

Optical depth contribution in path length ds is β (λ) ds

Mie Scattering



General Results

Size parameter x = 2 π r / λ for λ = wavelength

For very small particles x << 1 (Rayleigh scattering) scattering $\propto 1 / \lambda^4$ The sky is Blue

For medium size particles (aerosols), x ~1 (Mie scattering) scattering ~ $1/\lambda$ The sky is grey

For big size particles (cloud droplets, cirrus) x >> 1 Q_{ext} -> 2 independent of λ Clouds are white



Radiation detected by satellites



- Air scattering depends on geometry and can be calculated (Rayleigh scattering)
- Clouds completely obscure the surface and have to be masked out
 - Aerosols redirect incoming radiation by scattering and also absorb a fraction
 - Surface reflectance is a property of the surface

Satellite orbits

Advantages:

More near to Earth -> Higher spatial resolution Used also for Active Obs.(Radar/Lidar) and PMW

Disadvantages:

Poorer time resolution -> needs of constellation

<u>Advantages:</u> Better time resolution

Disadvantages:

One side of the Earth -> needs of constellations large viewing angles at the borders -> geometrical distorsions Only VIS/IR and passive Obs.





Satellite Detection of Aerosols

Just like the human eye, satellite sensors detect the total amount of solar radiation that is reflected from the earth's surface (R_o) and backscattered by the atmosphere from aerosol, pure air, and clouds. A simplified expression for the relative radiation detected by a satellite sensor (I/I_o) is:

$$I = \int_{Height} \int_{Type} \int_{Size} \int_{Angle} \int_{Shape} H \bullet C \bullet D \bullet P \bullet S \ dHdCdDdPdS$$

$$I / I_o = R_o e^{-\tau} + (1 - e^{-\tau}) P$$

where τ is the aerosol optical thickness and P the angular light scattering probability.

Today, geo-synchronous and polar orbiting satellites can detect different aspects of aerosols over the globe daily.





Apparent Surface Reflectance, R

• The surface reflectance R_0 is obscured by aerosol scattering and absorption before it reaches the sensor

• Aerosol acts as a *filter* of surface reflectance and as a *reflector* solar radiation



- The apparent reflectance , R, detected by the sensor is: $R = (R_0 + R_a) T_a$
- Under cloud-free conditions, the sensor receives the reflected radiation from surface and aerosols
- Both surface and aerosol signal varies independently in time and space
- Challenge: Separate the total received radiation into surface and aerosol components

Aerosol Effect on Surface Color and Surface Effect

- Aerosols add to the reflectance and sometimes reduce the reflectance of surface objects
- Aerosols always **diminish** the **contrast** between dark a bright surface objects
- Haze and smoke aerosols change the color of surface objects to bluish while dust adds a yellowish tint.



- **Dark** surfaces like ocean and dark vegetation makes the aerosol appear **bright**.
- **Bright** surfaces like sand and clouds makes the aerosol invisible.

Apparent Surface Reflectance, R

The critical parameter whether aerosols will increase or decrease the apparent reflectance, R, is the ratio of aerosol angular reflectance, P, to bi-directional surface reflectance, R_0 , P/ R_0

Aerosols will increase the apparent surface reflectance, R, if $P/R_0 < 1$. For this reason, the reflectance of ocean and dark vegetation increases with τ .

When $P/R_0 > 1$, aerosols will decrease the surface reflectance. Accordingly, the brightness of clouds is reduced by overlying aerosols.

At $P^{\sim} R_0$ the reflectance is unchanged by haze aerosols (e.g. soil and vegetation at 0.8 um).

At large τ (radiation equilibrium), both dark and bright surfaces asymptotically approach the 'aerosol reflectance', P





$$\tau = \frac{1}{m_0} \cdot \ln \frac{I_0}{I_{meas}}$$

This is valuable at a given wavelength λ

Image is measured, Io und mo are known.

This shows that the measurement of Immi is sufficient to invert the Optical Depth (t).

The Optical Depth t quantifies the turbidity of the atmosphere. The optical depth of the atmosphere is the sum of the optical depth of all atmospheric components separately:

$$\tau = \tau_R + \tau_{aer} + \tau_{H_20} + \tau_{0_3} + \tau_{ag}$$

Photometers – ground-based

Methodology for the inversion of AOD, PWV und O₃DU

The values of $\tau_{\alpha,g}$, τ_{H_20} and τ_{0_3} are 0 for all wavelength λ , for which there are no line of gas absorption (see section "Spectroscopy and UV radiation"). It is easy to invert the AOD ($\tau_{\alpha ev}$) throw a photometrical measurement in a spectral channel where the values of $\tau_{\alpha,g}$, τ_{H_20} and τ_{0_3} are 0. The AOD is therefore estimated with a measurement at these wavelengths, and then the AOD values for other wavelengths are extrapolated using the law of Ångström.

We invert τ_{0_3} thanks a photometrical measurement in ultraviolet (UV), where the values of all optical depths are 0 except the Rayleigh one, the ozone one and the AOD (this is possible for several wavelength λ).

We invert r_{H_20} thanks to a photometrical measurement in near infrared (NIR), at wavelengths where the values of all optical depths are 0, except the Rayleigh one, the one of water vapor and the AOD.

The water vapor column PWV (in mm) and the ozone column (in Dobson Unit: DU) are inverted from the values of r_{H_20} and r_{0_3} respectively thanks to the tabulated values of the absorption coefficient of these gases.

Climatology from AERONET



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Active Remote Sensing - Lidar



Active Remote Sensing – Lidar Satellite



Advantages and drawbacks – my personal view

Figure 3: Air pollution monitoring in Africa







Advantages and drawbacks – my personal view

In-situ gravimetric	Measurements of mass – accurate	Local coverage – no over sea – relatively expensive – no aerosol types
In-situ optical	Cheaper, potentially pervasive	Not direct, less accurate, calibration needed
Remote Sensing satellite	Global, cover long time range	Optical parameters, difficult to derive aerosol type – no info on vertical
Ground-based	Dense networks, calibrated, insights on aerosol size	Not global, no info on vertical
Lidar	Information on vertical structure, insights on aerosol size and optical properties	Not global, expensive

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Few "historical papers"

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