Lesson 6:
ATMOSPHERIC AEROSOLS

Course: Laboratory of Atmospheric Remote Sensing
Laurea Magistrale in Atmospheric Science and Technology

Contents (1)

- Atmospheric aerosol: definition, hystorical importance, properties, classification criteria
- Residence time
- Aerosol size distribution: nucleation mode, accumulation mode, coarse mode
- Aerosols and visibility: light scattering by aerosols
- Determination of aerosol particle size: particle collection efficiency
- Aerosols and properties of interest: AOD, AE, SSA
- Tropospheric aerosol: sources, chemical properties, transport, sink, particle surface area distribution, formation and removal processes
- Aerosol and health, aerosol and water, aerosols and plants

Contents (2)

- Radiative forcing
- Stratospheric aerosols: unperturbed stratospheric ozone, remote sensing measurements of O3, the ozone hole, sulfur in the stratosphere
- Urban aerosols: urban aerosol size distribution, PM2.5 and PM10, vertical distribution of urban aerosol
- Saharan dust outbreaks
- The HYSPLIT model: saharan dust outbreak, fires and advection of continental air masses

Reading material:

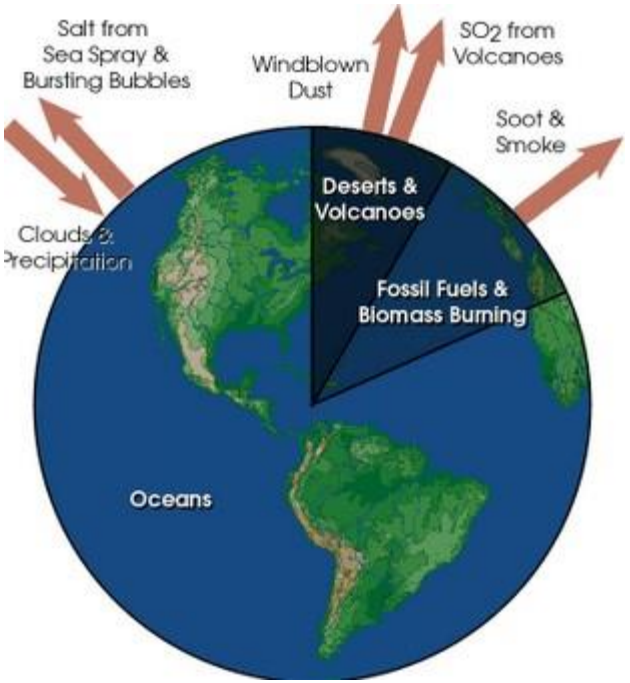
Wallace & Hobbes, Atmospheric Science: an introduction survey. Chps. 4-5.

What is Atmospheric Aerosol?

Aerosols are small solid and/or liquid particles suspended in the atmosphere with negligible terminal fall speeds. When these particles are sufficiently large, we notice their presence as they scatter and absorb sunlight.
Their scattering of sunlight can reduce visibility (haze) and redden sunrises and sunsets.

Aerosols interact both directly and indirectly with the Earth's **radiation budget and climate**.

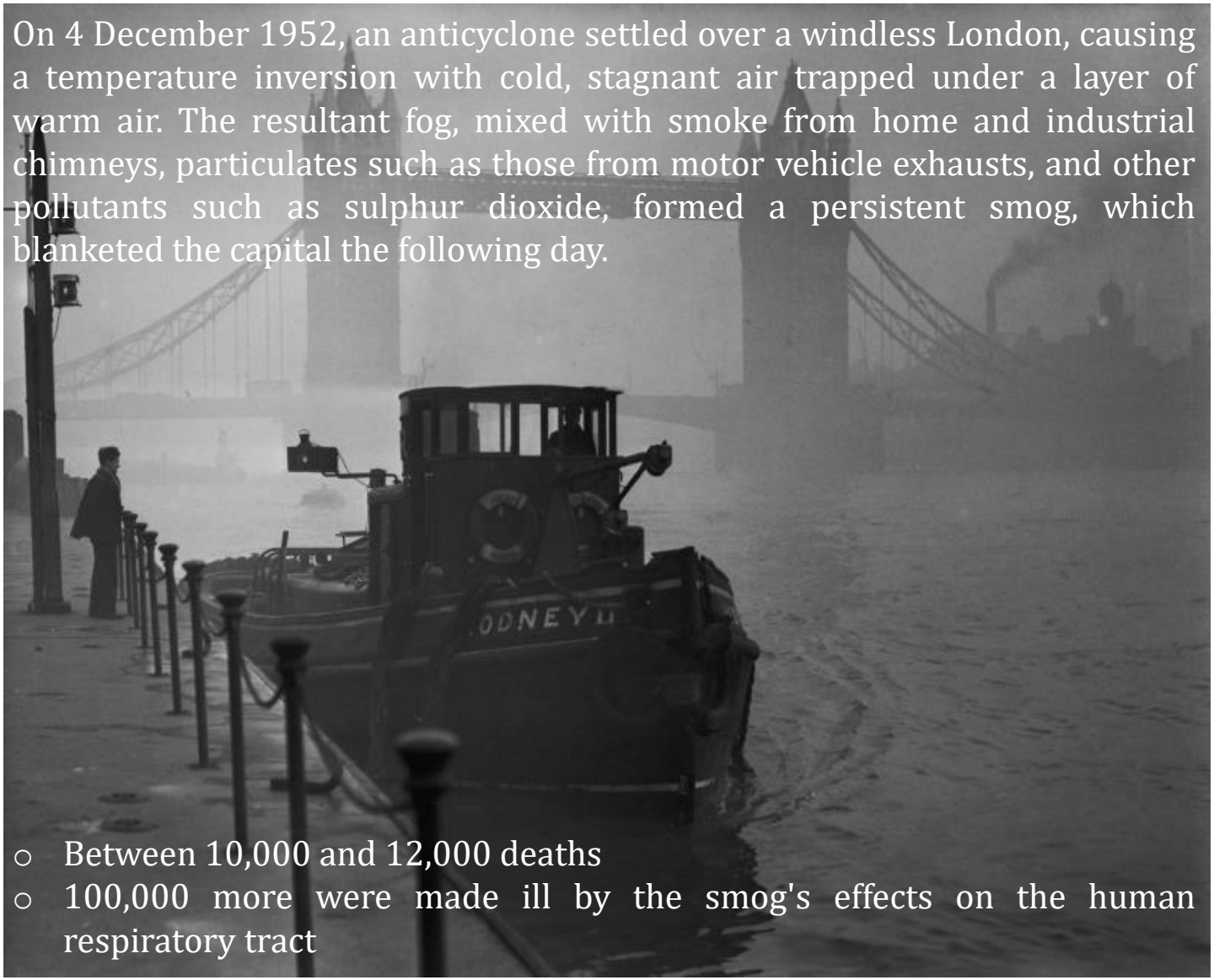
As a **direct effect**, the aerosols scatter sunlight directly back into space.
As an **indirect effect**, aerosols in the lower atmosphere can modify the size of cloud particles, changing how the clouds reflect and absorb sunlight, thereby affecting the Earth's energy budget.



Hystorical importance (1)

Great Smog of London (1952)

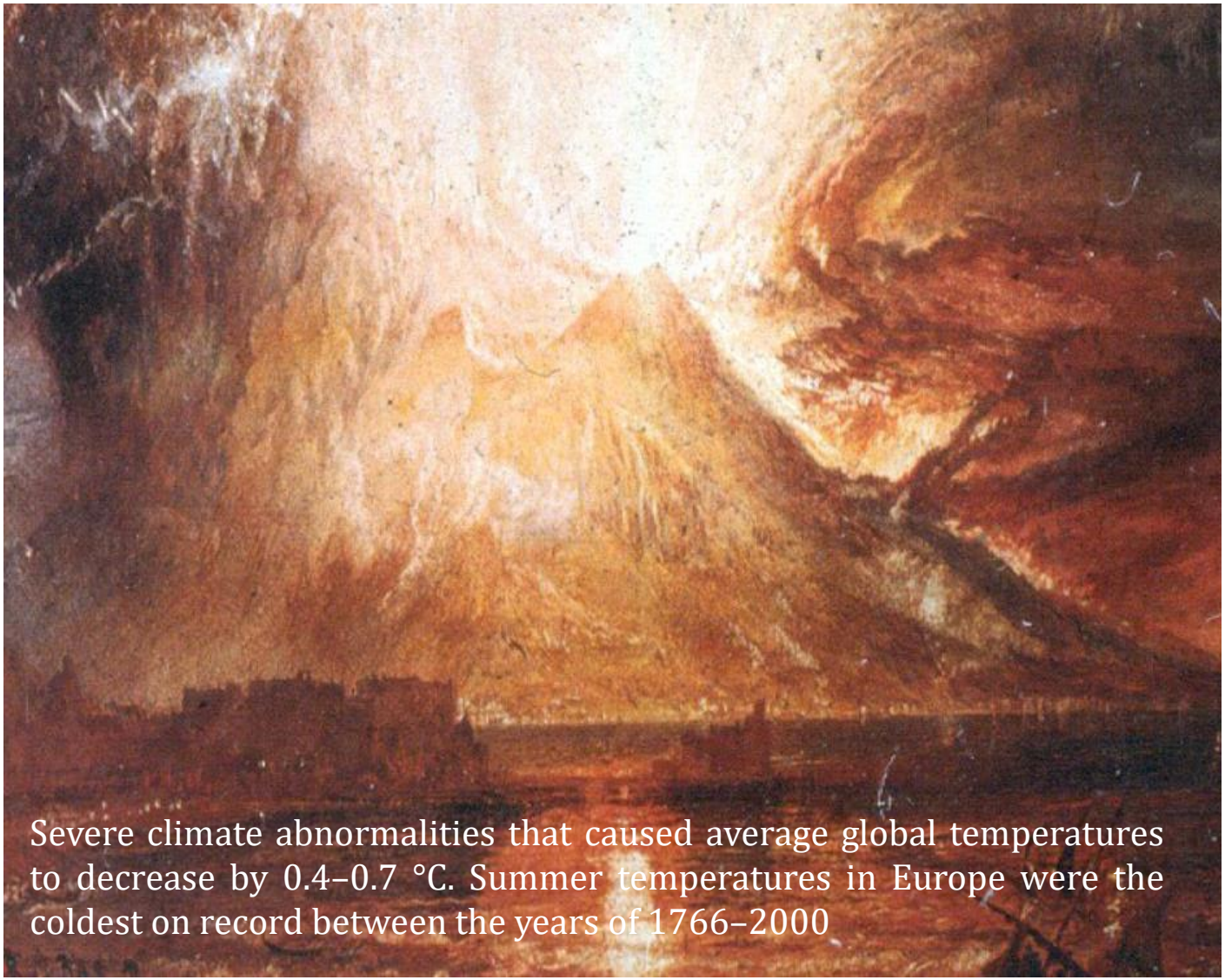
On 4 December 1952, an anticyclone settled over a windless London, causing a temperature inversion with cold, stagnant air trapped under a layer of warm air. The resultant fog, mixed with smoke from home and industrial chimneys, particulates such as those from motor vehicle exhausts, and other pollutants such as sulphur dioxide, formed a persistent smog, which blanketed the capital the following day.



- Between 10,000 and 12,000 deaths
- 100,000 more were made ill by the smog's effects on the human respiratory tract

Hystorical importance (2)

Year Without a Summer (1816)



Severe climate abnormalities that caused average global temperatures to decrease by 0.4–0.7 °C. Summer temperatures in Europe were the coldest on record between the years of 1766–2000

Hystorical importance (3)

Year Without a Summer (1816)

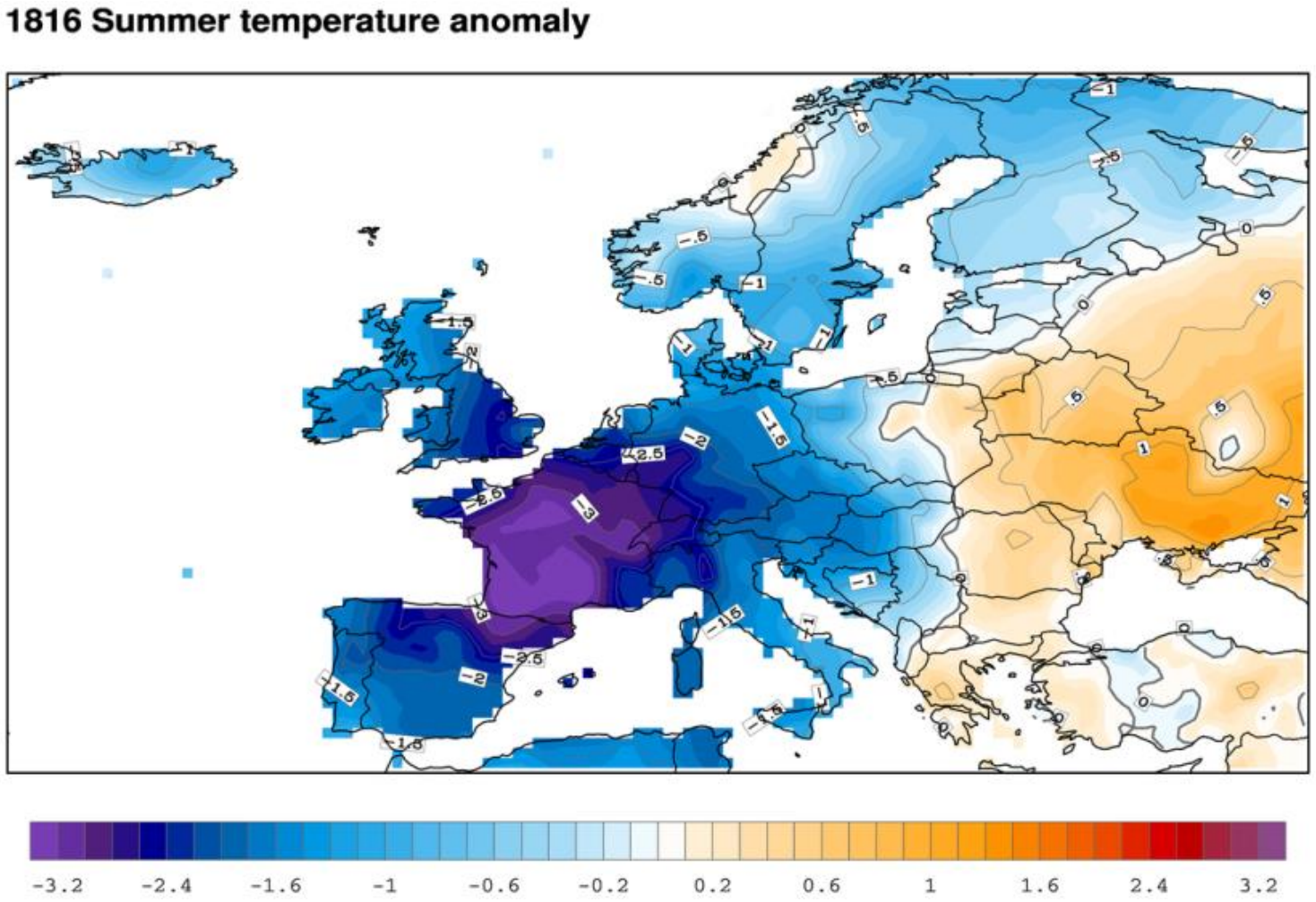
In Asia, the cold weather and flooding killed trees, rice crops and even water buffalo.
In Europe, heavy rains and cool temperatures throughout Britain and Ireland caused reduced or eliminated the harvest and famine was wide-spread.
In the spring and summer of 1816, a persistent "dry fog" was observed in parts of the eastern United States. Some cities in New England saw snow fall as late as June 6th.

Several volcanic eruptions characterized the period: Mount Tambora (Indonesia) 1815, the southwestern Pacific Ocean (1808), Caribbean (1812), Dutch East Indies (1812), Japan (1813), Philippines (1813)

These eruptions had built up a substantial amount of atmospheric dust (at least 100 km^3 only from Mount Tambora).
As is common after a massive volcanic eruption, temperatures fell worldwide because less sunlight passed through the stratosphere.

Hystorical importance (4)

Year Without a Summer (1816)



Aerosols classification criteria

Dimensions:

r < 0.1 mm	nucleation mode (Aitken nuclei)
0.1< r < 1 mm	large particles (accumulation mode)
r > 1 mm	giant particles (coarse mode)

Location:

Tropospheric, Stratospheric, Marine, Continental, Arctic

Source:

Natural, Anthropogenic, Volcanic

Process:

Primary, Secondary

Composition:

Powders (from the ground or industrial), Sea salt, Carbonaceous (organic and non-organic), Biological, Sulphates, Nitrates.

Criterion adopted by the IPCC 2001 (Intergovernmental Panel on Climate Change)

Residence time (1)

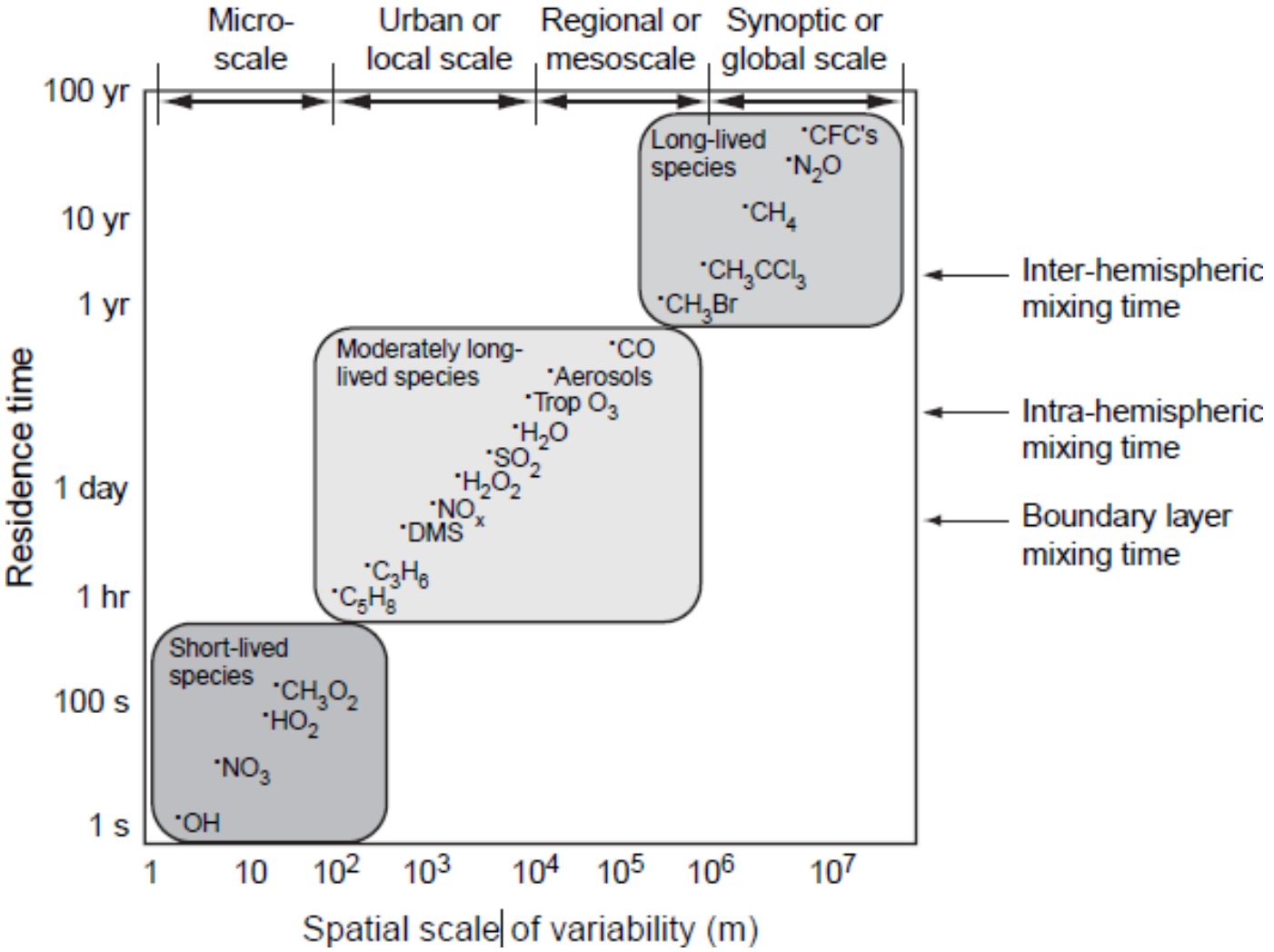
The globally averaged concentration of a trace constituent in the atmosphere does not change significantly over a given time period.
The rate at which the constituent is injected into (and/or produced within) the atmosphere must equal the rate at which it is removed from the atmosphere.

Under such steady-state conditions we can define the **residence time** (or **lifetime**) τ **of a trace constituent in the atmosphere** as:

$$\tau = \frac{M}{F}$$

M (kg) = amount of the constituent in the atmosphere
F (kg s^{-1}) = rate of constituent removal from the atmosphere.

Residence time (2)



Residence time does not always give a representative idea of how long it would take for the atmospheric concentration of a species to react to an abrupt change in the source.

Residence time (3)

Residence times may be determined by:

(i) **physical removal processes** (e.g. scavenging by precipitation)

(ii) **chemical processes**

If a chemical species has a very short (or very long) residence time in the atmosphere, significant variations in the concentration of the species will generally occur over very short (or very large) spatial scales.

Species with short residence times will be present in high concentrations close to localized sources and in low concentrations far removed from their sources.

In contrast, chemical species with long residence times exhibit more uniform concentrations.

Aerosols size distribution (1)

0.003 - 100 μm

minimum measurable size

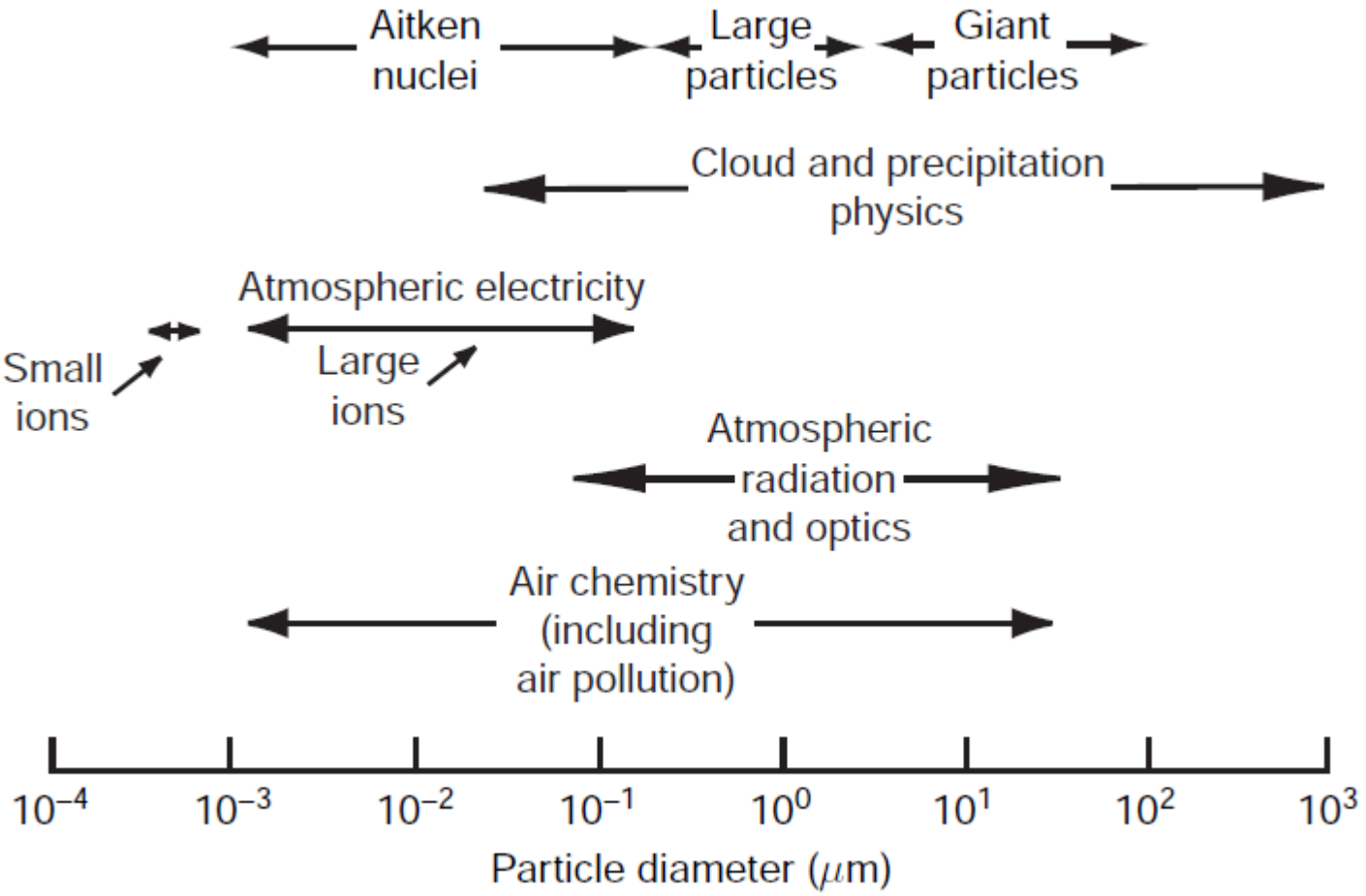
larger particles quickly fall to the ground

Aerosols can be emitted in particulate form as:

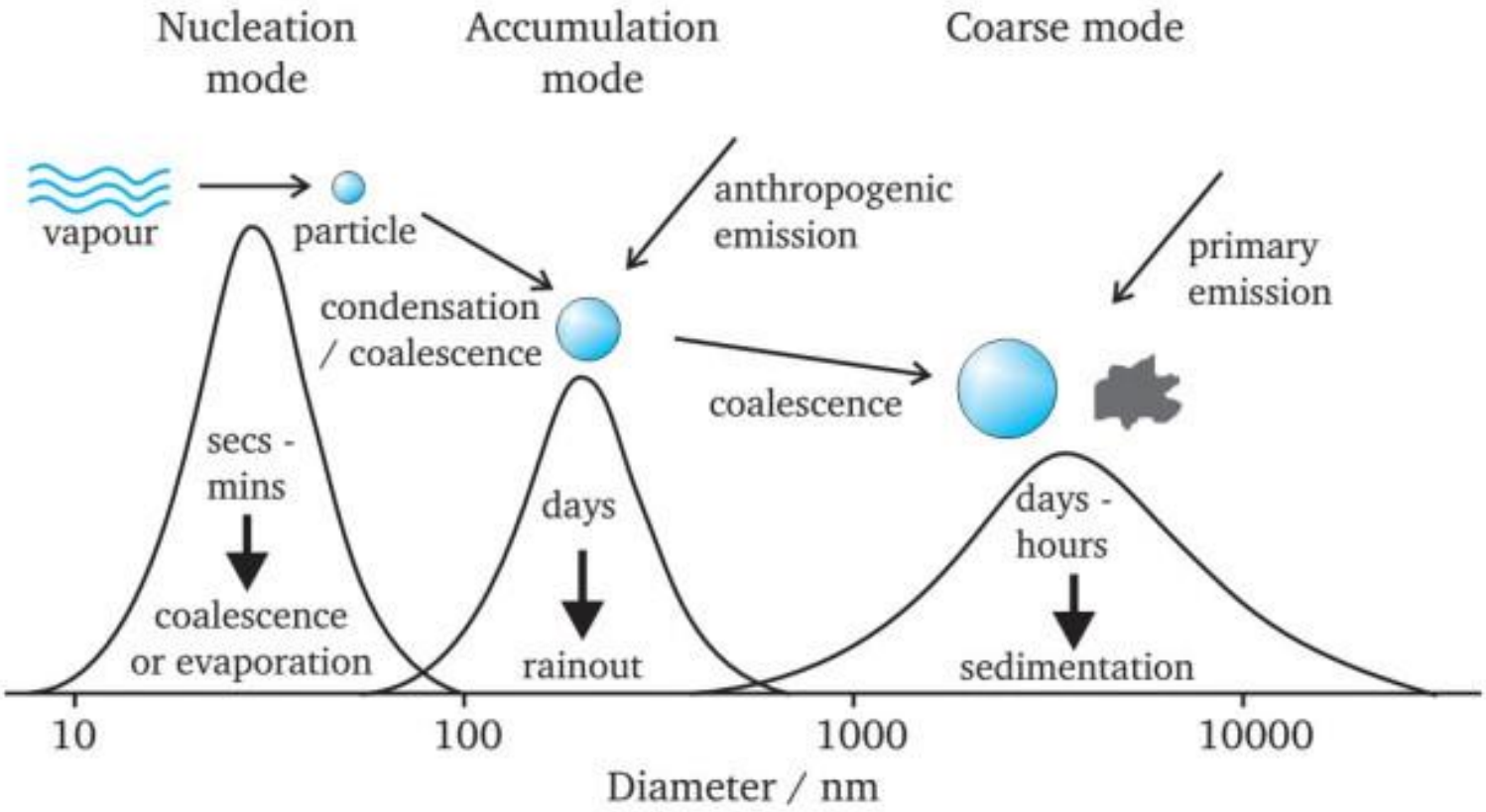
- **primary aerosols**: directly into the atmosphere, producing particles of all sizes

- **secondary aerosols**: formed in the atmosphere by gas-to-particle conversion involving chemical reactions of gases that result in condensable species with particles in the nanometer size range (<0.1 μm) with very short lifetimes. In fact, they rapidly become attached to larger particles and can exist in the atmosphere at significant concentrations only if they are continuously produced.

Aerosols size distribution (2)



Aerosols size distribution (3)



The lifetime of an aerosol in the atmosphere ranges from minutes to days or weeks and they may be lost from the atmosphere due to complete evaporation, gravitational sedimentation, impaction or rain-out.

Nucleation mode (Aitken nuclei)

The smallest size range, composed of **aerosols < 0.1 μm**, is called the **Aitken nuclei** (or **nucleation mode**).

The existence of these aerosols was discovered in 1875 by Coulier and were thoroughly studied for the first time by Aitken at the turn of the century.

These ultrafine aerosols are produced by ambient-temperature gas-to-particle conversion and also by combustion processes generating hot gases that subsequently undergo condensation.

Aerosols in the Aitken range have particle diffusion coefficients > 0.001 cm^2/sec and they can diffuse rapidly to the surfaces of other particles. In addition, these ultrafine aerosols can act as nuclei for the condensation of low-vapor-pressure gases causing them to grow rapidly into the next size range.

Accumulation mode (coarse particles)

Aerosols in the intermediate size range from approximately **0.1-2 μm** are known as the **accumulation range**. They are so named because the particle removal mechanisms of sedimentation and diffusion to surfaces are least effective in this range causing particles of this size to accumulate in the atmosphere. Aerosols in this fine size range typically arise from coagulation of smaller particles in the Aitken range or from condensation of low-volatility gases (water vapor, organics, etc.) onto existing particles.

Because of their sources, particles in the accumulation range typically contain organic compounds and soluble inorganics such as ammonium (NH_4^-), nitrate ($N0_3^-$), and sulfate ($S0_4^{2-}$). Being too small to settle out of the atmosphere, they are removed relatively slowly, primarily by incorporation into clouds and subsequent rainout. The rate of cloud droplet formation from these aerosols depends on their chemical compositions and their hygroscopicity, with the more soluble species being removed faster. Alternately, fine aerosols can be removed from the atmosphere by dry deposition after being carried to surfaces by eddy diffusion.

Coarse mode (giant particles)

Atmospheric aerosols in the **coarse range**, **larger than approximately 2 μm**, are usually produced by mechanical processes such as grinding or wind erosion.

Thus, because of the nature of their sources, they are composed predominately of minerals and inorganics such as sand and sea salt. Also included in this size range are larger bioaerosols such as spores, pollen, and bacteria.

With settling velocities >0.01 cm/sec, coarse aerosols are generally removed from the atmosphere fairly rapidly by sedimentation. However, the atmospheric transport of coarse aerosols can occur over relatively long distances by convective processes where fallout is balanced by re-entrainment. E.g., mineral dusts from western China have been detected in western North America and Canada and African mineral dusts have been detected in south central Florida .

Aerosols and visibility

Light scattering by atmospheric aerosols can:

- reduce the perceived contrast of distant objects by deflecting the light from the object out of the line of sight
- during the daytime, sunlight is simultaneously scattered into the line of sight, making dark objects appear lighter

The absorption of light by gas molecules and particles can:

- cause the change of color in the atmosphere
- reduce atmosphere visibility

Light scattering by aerosols (1)

Light scattering by particles depends on the size of the particles relative to the wavelength *(λ)* of light.

1) For particle diameters much less than λ *(**D*<**0.03 µm** for the UV/visible region), light scattering is similar to that by gaseous molecules (**Rayleigh scattering**).

The scattered intensity is symmetrical in all directions relative to the incident light and varies as λ^{-4}.

2) For particles much larger than λ *(**D*** **>10µm**), **light is scattered by reflection** from the surface, **diffraction** around the edges, or **refraction** through the interior of the particle (geometric scattering).

Most of the light scattered in this manner is not diverted significantly from its original path.

Light scattering by aerosols (2)

3) Particles in the intermediate size range ***(D=λ)*** scatter light most efficiently and in a more complicated manner, known as **Mie scattering**.

The intensity of light (I) scattered at distance r and scattering angle θ from a particle is given by

$$I(\theta, r) = \frac{I_0 \lambda^2 (i_1 + i_2)}{8\pi^2 r^2}$$

where

I_0 = incident light intensity

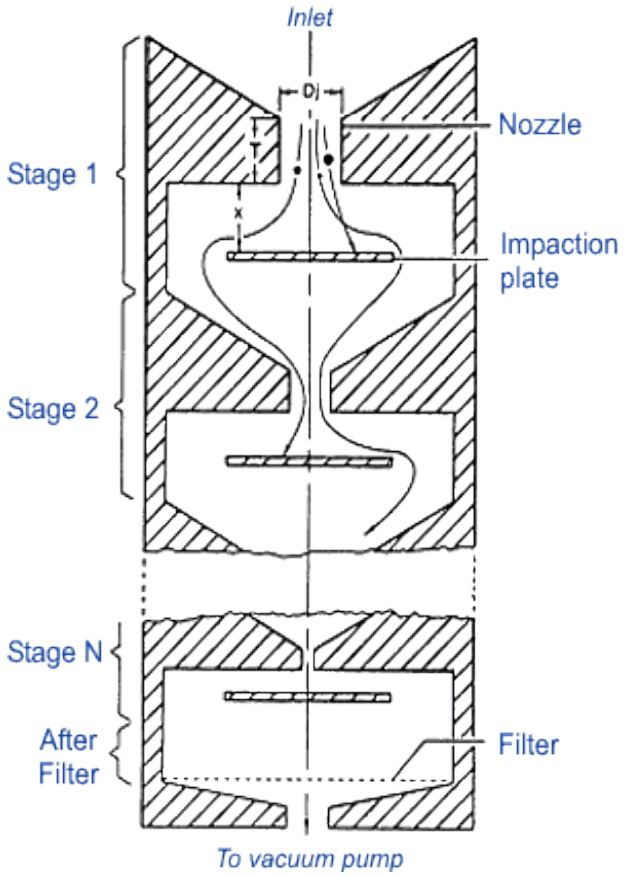
$i_1, i_2 =$ Mie intensity parameters for the perpendicular and parallel polarized components of the scattered light

The Mie scattering parameters are a complex function of the refractive index (m) and the size parameter ($\alpha = \pi D/\lambda$) of the particle and the scattering angle (θ).

How to determine particles size?

Since atmospheric particles are rarely spherical, their size is typically described in terms of an **effective aerodynamic diameter**, defined as the diameter of a sphere of unit density (1 g/cm3) that has the same settling velocity as the atmospheric particles.

The size ranges of atmospheric aerosols are typically determined by using **impactors**: when an airstream is bent sharply, the particles in the air tend to continue in a straight line because of their inertia. In an **multiple stage (Lundgren-type cascade) impactor** a collection plate placed in an air flow causes the gas to stream around the plate. However, particles in the air may strike the plate and stick to it, depending on the size of the particle and the velocity of the air stream.

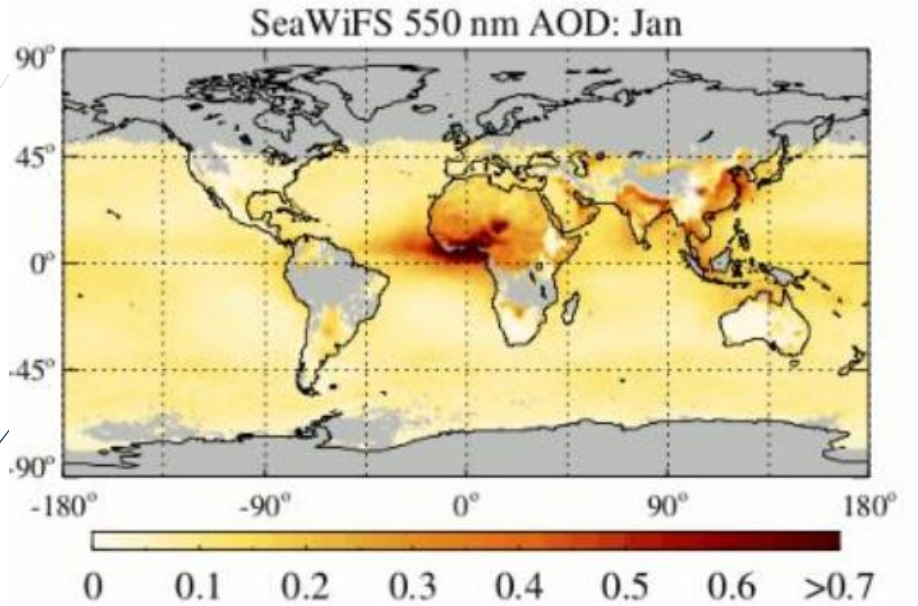


Particle collection efficiency

$$\eta = \frac{D^2 V \rho}{18 \mu D_b}$$

η = particle collection efficiency
D = particle diameter
V = flow velocity of the air
ρ = particle density
μ = viscosity of air
D_b= curvature of the air stream, which depends on the physical dimensions of the impactor

Aerosols and properties of interest: AOD (1)



Aerosol Optical Depth (AOD): total column optical extiction of aerosol at a given wavelength

- Most commonly, 550 nm
- Related to how much aerosol is in the atmosphere
- Also termed aerosol optical thickness (AOT)

Aerosols and properties of interest: AOD (2)

«Particles in the atmopshere (dust, smoke, pollution) can block sunlight by absorbing or by scattering light. AOD tells us how much direct sunlight in prevented from reaching the ground by these aerosol particles».

Provençal et al., 2017

Higher AOD (e.g. 1) = Hazy
Lower AOD (e.g. 0,1 or lower) = Clear sky

Aerosols can travel a long way

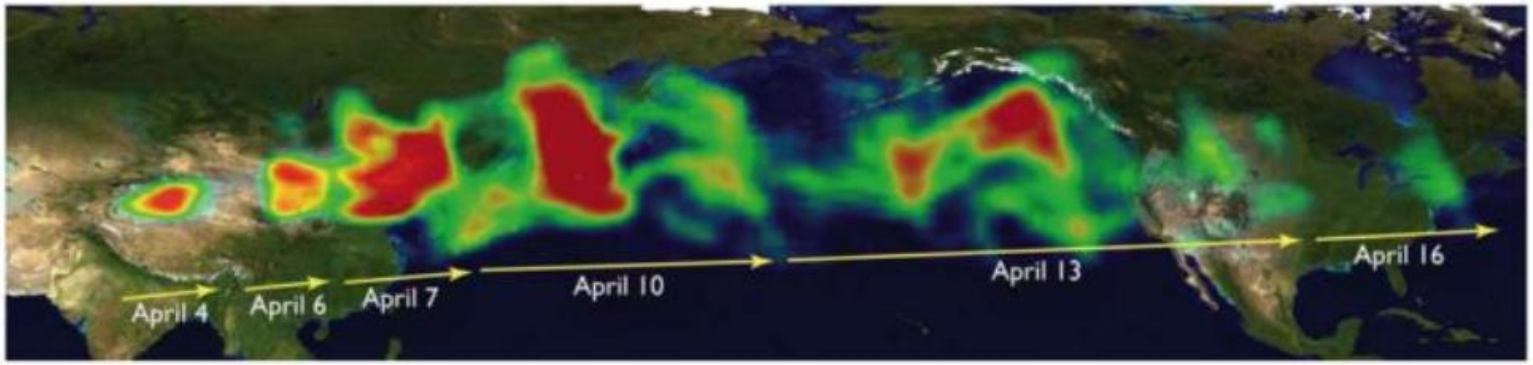
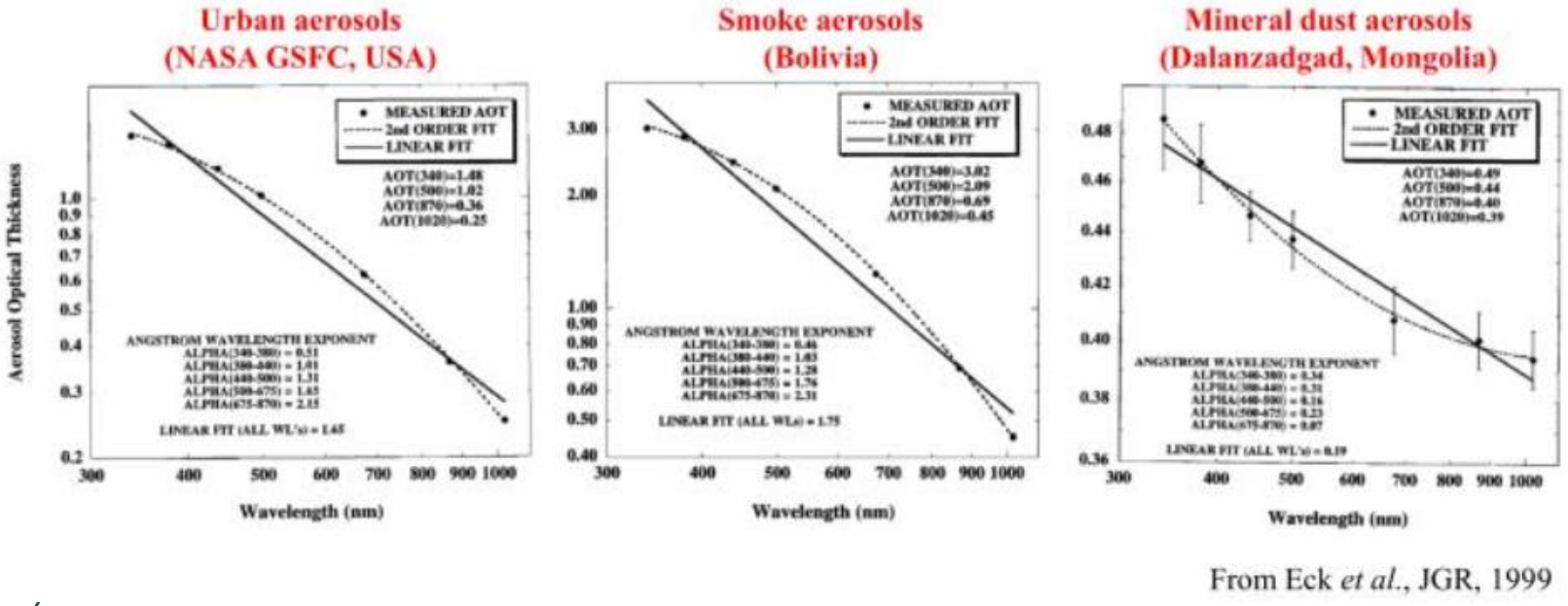


Fig. 1. Time series of TOMS AI composite in April 2001 showing the long-range transport of Asian dust across the Pacific reaching as far as the east coast of the U.S.

- Mineral dust from Taklimakan desert transported to North America
- We also see Asian pollution transported to North America, Saharan dust transported to the Amazon and Europe, high-latitude smoke circling the world in both hemisphere, etc..

Aerosols and properties of interest: AE

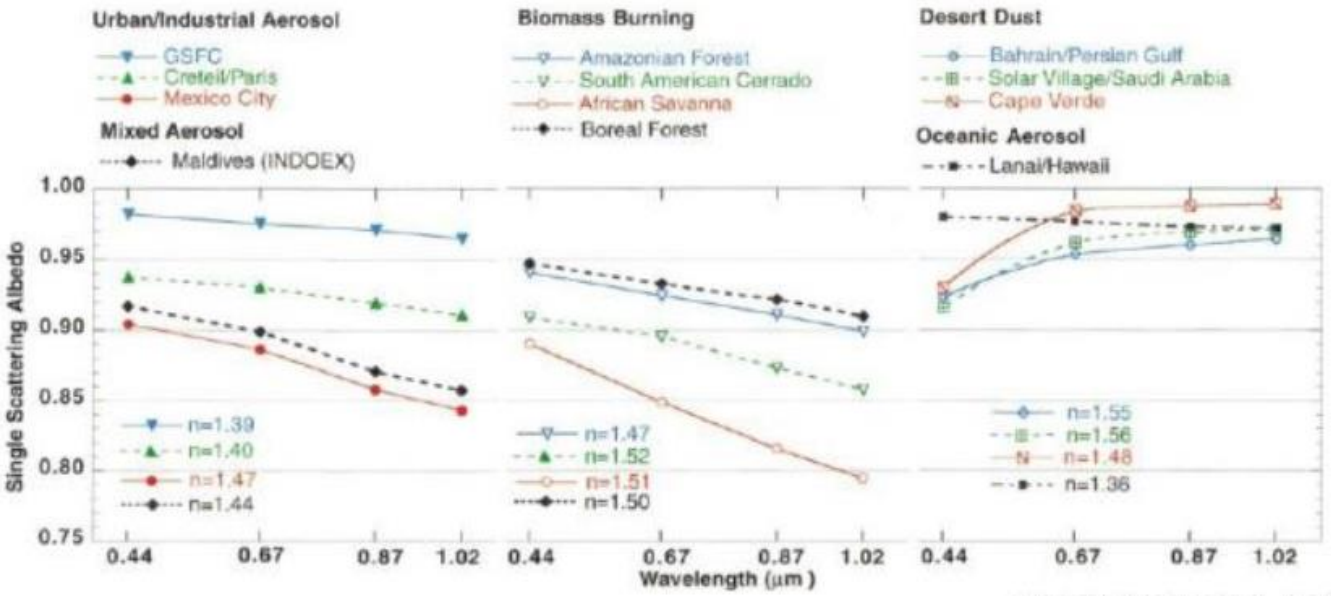


From Eck *et al.*, JGR, 1999

Ångström exponent (AE, α): spectral dependemnce of AOD

- AE<1 suggest optical dominance of coarse particle (e.g. dust)
- AE>1 suggest optical dominance of fine particle (e.g. smoke)
- Depends on wavelenght range used to calculate it

Aerosols and properties of interest: SSA



From Dubovik *et al.*, JAS, 2002

Single scatter albedo (SSA): measure of light absorption by aerosols

- SSA=0 pure absorbing aerosols (never encountered)
- SSA=1 pure scattering aerosols
- Typical range: ≈0,8 (industrial, smoke) – 0,99 (continental, marine)
- Also has wavelenght dependence

TROPOSPHERIC AEROSOLS

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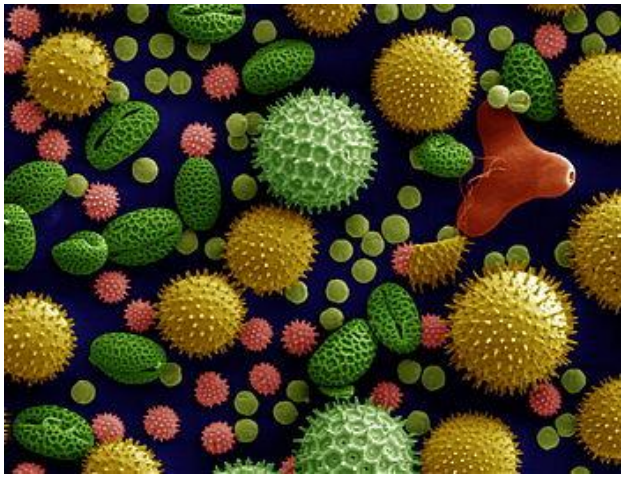
Tropospheric aerosols – sources (1)

(1) Biological

Solid and liquid particles are released into the atmosphere from **plants** and **animals** (seeds, pollen, spores, and fragments of animals and plants), usually 1–250 μm in diameter.

Bacteria, **algae**, **protozoa**, **fungi**, and **viruses** are generally 1 μm in diameter.

Small smoke particles from **forest fires** (primarily organic compounds and elemental carbon) and fly ash are injected directly into the air (peak at 0.1 μm diameter).



Tropospheric aerosols – sources (2)

(1) Biological

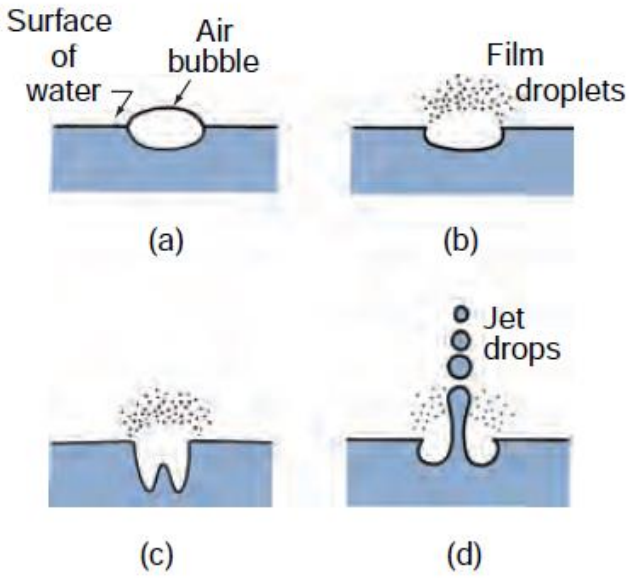
The oceans eject materials into the air is bubble bursting :

a) some materials enter the air in drops torn from windblown spray and foam. Aerosols composed of sea salt originate from droplets ejected into the air when air bubbles burst at the ocean surface.

b) Many small droplets are produced when the upper portion of an air bubble film bursts; these are called film droplets.

c) After evaporation, the film droplets leave behind sea-salt particles with diameters less than 0.3 μm.

d) From one to five larger drops break away from each jet and these jet drops are thrown about 15 cm up into the air. Some of these drops subsequently evaporate and leave behind sea-salt particles with diameters >2 μm.



Tropospheric aerosols – sources (3)

(2) Solid Earth

The transfer of particles to the atmosphere from the Earth's surface is caused by **winds** and **atmospheric turbulence**.
The threshold values are at least 0.2 m s^{-1} for particles 50–200 μm in diameter.
Volcanoes inject gases and particles into the atmosphere. The large particles have short residence times, but the small particles can be transported globally, particularly if they reach high altitudes.

Tropospheric aerosols – sources (4)

(2) Solid Earth

Soil erosion causes **dust** and **sand storms** that put PM into air.
Cities with high PM have a higher death rate.
Dust also carries bacteria, viruses, fungi, pesticides, herbicides and heavy metals.

Tropospheric aerosols – sources (5)

(3) Anthropogenic

The global input of particles into the atmosphere from anthropogenic activities is 20% (by mass) of that from natural sources. The main anthropogenic sources of aerosols are **dust from roads**, **wind erosion** of tilled land, **biomass burning**, **fuel combustion**, and **industrial processes**.

Tropospheric aerosols – sources (6)

(3) Anthropogenic

EPA found indoor concentrations of toxic air pollutants are often higher than outdoor.
Organic fuels (wood) make up majority of household energy in developing countries.
These are often burned in smoky, poorly ventilated heating and cooking fires.

Tropospheric aerosols – sources (7)

(4) In situ formation

Gases may condense onto existing particles, thereby increasing the mass (but not the number) of particles, or gases may condense to form new particles. The former path is favored when the surface area of existing particles is high and the supersaturation of the gases is low.

Three major families of chemical species are involved in condensation of gases: **sulfur**, **nitrogen**, and **organic and carbonaceous** materials.

Tropospheric aerosols – sources (8)

(a) Direct emissions

	Northern hemisphere	Southern hemisphere
Carbonaceous aerosols		
Organic matter (0–2 μm)[a]		
Biomass burning	28	26
Fossil fuel	28	0.4
Biogenic (>1 μm)	—	—
Black carbon (0–2 μm)		
Biomass burning	2.9	2.7
Fossil fuel	6.5	0.1
Aircraft	0.005	0.0004
Industrial dust, etc. (>1 μm)		
Sea salt		
<1 μm	23	31
1–16 μm	1,420	1,870
Total	1,440	1,900
Mineral (soil) dust		
<1 μm	90	17
1–2 μm	240	50
2–20 μm	1,470	282
Total	1,800	349

(b) *In situ*

	Northern hemisphere	Southern hemisphere
Sulfates (as NH_4HSO_4)	145	55
Anthropogenic	106	15
Biogenic	25	32
Volcanic	14	7
Nitrate (as NO_3^-)		
Anthropogenic	12.4	1.8
Natural	2.2	1.7
Organic compounds		
Anthropogenic	0.15	0.45
Biogenic	8.2	7.4

[a] Sizes refer to diameters. [Adapted from Intergovernmental Panel on Climate Change, *Climate Change 2001*, Cambridge University Press, pp. 297 and 301, 2001.]

Chemical properties

- The large particles (coarse range) are generated by mechanical processes such as grinding and wind erosion. These particles, therefore, are generally composed of soil elements (Si, Al, Fe, Mg, Ca, Na, K, etc.) from the continental materials and sea salt elements (Na, CI, Mg, Ca, K, etc.) from the oceans.

- Fine particulate matter (< 2.5 μm) is generally formed by gas-to-particle conversion and combustion processes. The major components are $S0_4^{2-}$, $N0_3^-$, NH_4^+, elemental and organic carbon, along with trace elements from combustion processes.

Transport

The transport can be over intercontinental, even global, scales.
The aerosols produced by gas-to-particle conversion, can be transported longrange because the time required for gas-to-particle conversion and the relatively small sizes of the particles produced by this process lead to long residence times in the atmosphere.

Example: sulfates from SO_2 blasted into the stratosphere by large volcanic eruptions.

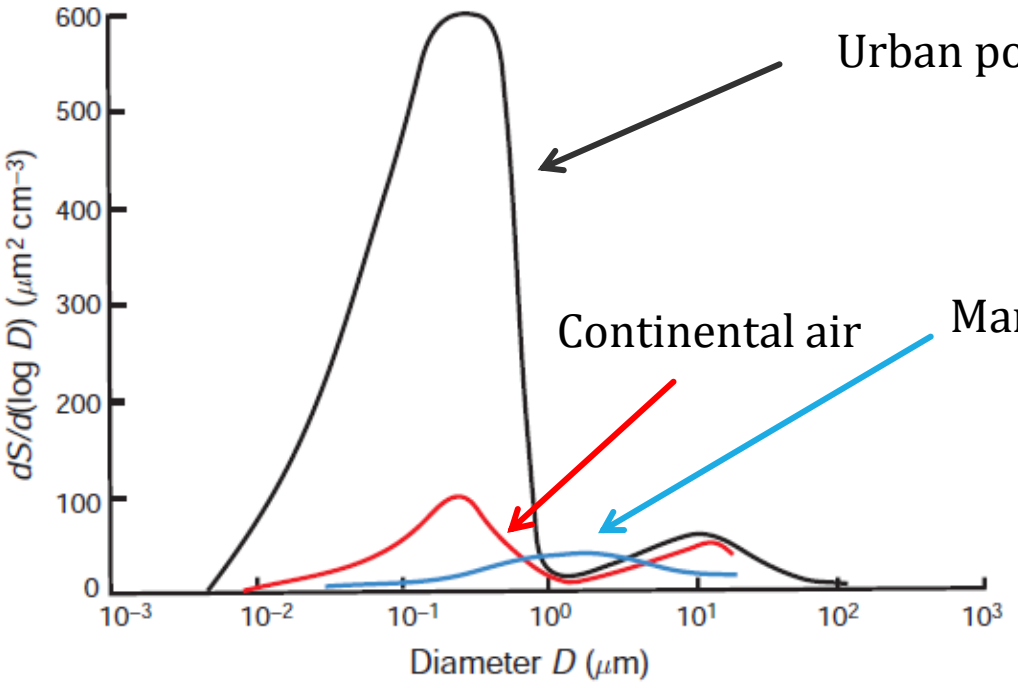
Example: acidic aerosols such as sulfates and nitrates, which contribute to acid rain.

Sink

On average, particles are removed from the atmosphere at about the same rate as they enter it.

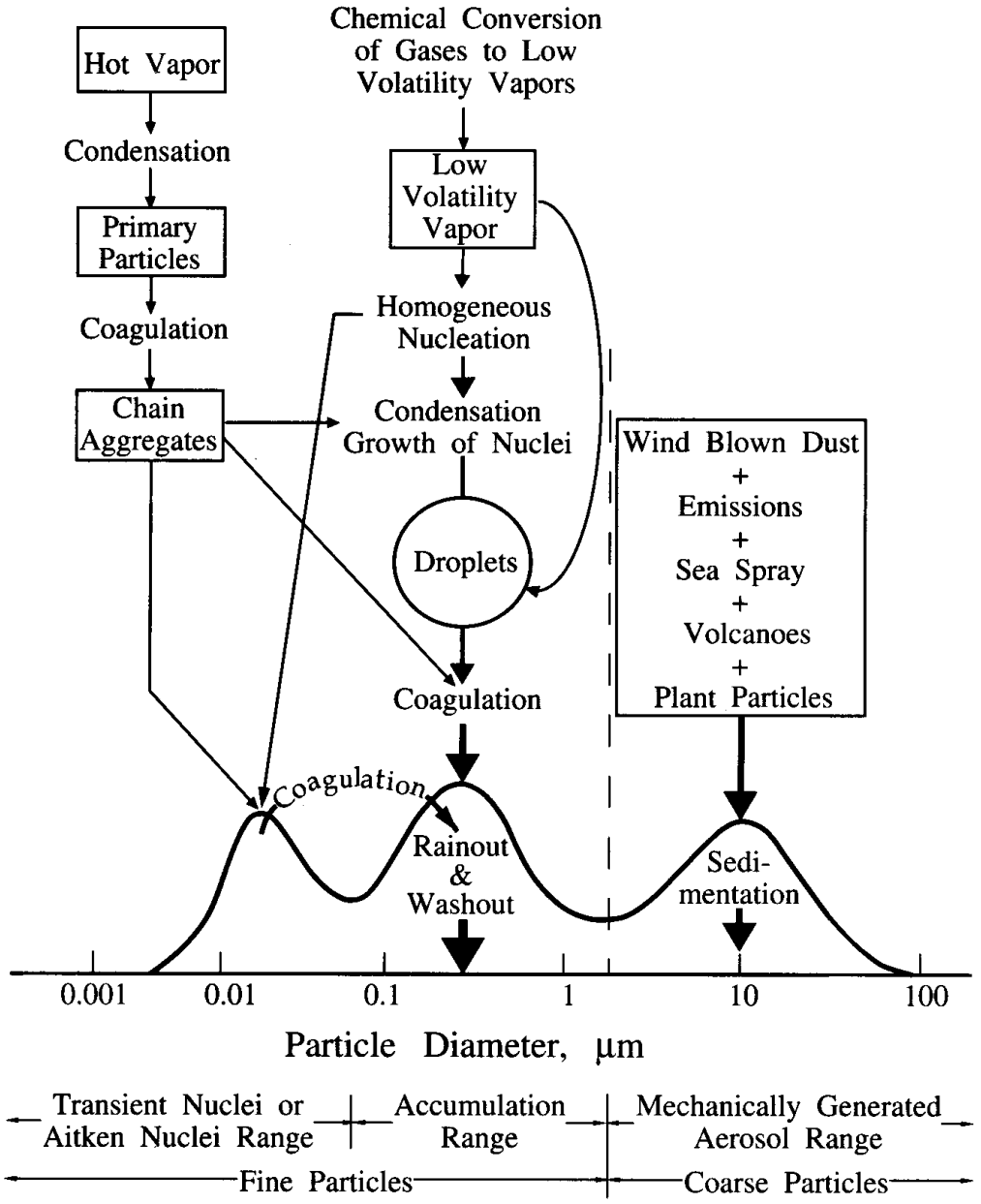
Small particles can be converted into larger particles by coagulation. Because the mobility of a particle decreases rapidly as it increases in size, coagulation is essentially confined to particles less than 0.2 μm in diameter. Although coagulation does not remove particles from the atmosphere, it modifies their size spectra and shifts small particles into size ranges where they can be removed by other mechanisms.

Particle surface area distributions



Desig-nation	Aitken nuclei	Large particles	Giant particles
Sources	← Combustion → ← Gas-to-particle conversions → ← Fly ash, sea-salt, pollens → ← Windblown dusts → ← Coagulation of Aitken nuclei → ← Cloud droplet evaporation → ← Giant particles from industries →		
Sinks	← Coagulation → ← Capture by cloud particles → ← Precipitation scavenging ← Dry fallout →		
Resi-dence time	Less than an hour in polluted air or in clouds	Days to weeks	Hours to days	Minutes to hours

Formation and removal processes



Differences in aerosols

Aerosol type	Number density (N/cm^3)	Mass concentration ($\mu g/m^3$)
Industrial plume	10^7	10^2-10^5
Urban area	10^5-10^6	100-1000
Rural area	10^3-10^5	10-100
Remote Area	100-500	10
Arctic	10-1000	0.1-10
Antarctic	10-100	0.1

Aerosol and health

WHO estimates each year 5-6 million people die prematurely from illnesses related to air pollution.

Likelihood of suffering ill health is related to intensity and duration of exposure.

As much as a 5 to 10 year decrease in life expectancy if you live in worst parts of Los Angeles.

Particulates less than 2.5 micron have been linked with heart attack, asthma, lung cancer and abnormal fetal development.

New rules will remove PM from diesel engines and power plants.

Most air pollutants are inhaled, but some can be directly absorbed through the skin or ingested in food and water.

Human Health effects (1)

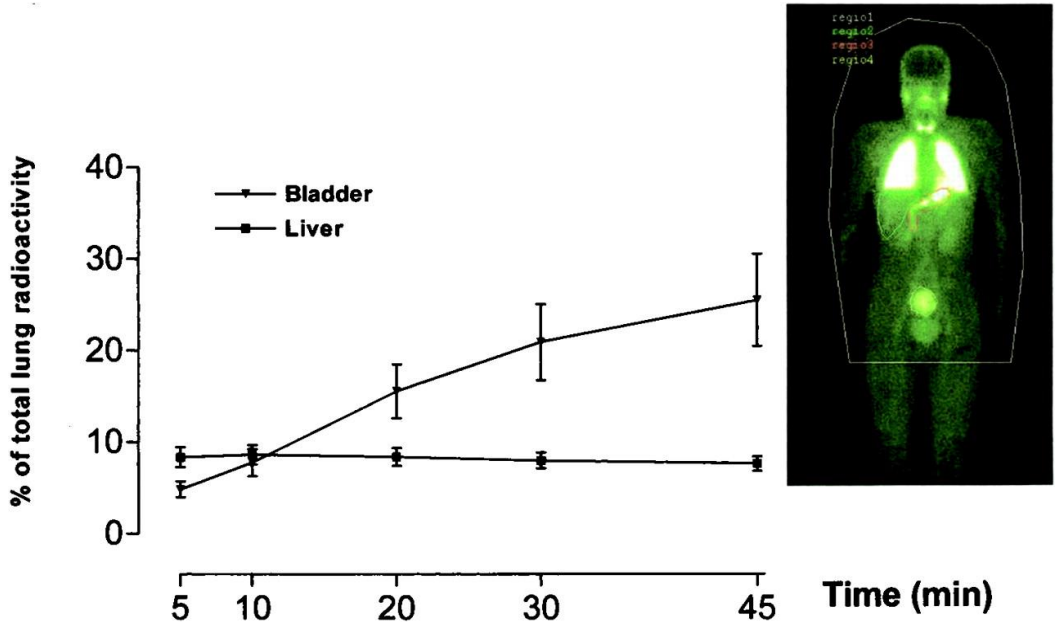
Table II. Factors that influence particulate deposition in the human respiratory system (*21*).

Particle Size (μm)	*Area Deposited*	*Method*	*Air Direction Change*	*Relative Air Velocity*
5–30	Nasopharyngeal	Impaction	Very abrupt	4
1–5	Tracheobronchial	Sedimentation	Less abrupt	3–2
< 1	Pulmonary	Diffusion	Calm	0–1

Ultrafine particles and nanoparticles < 0.1 μm are deposited in the deep lung by diffusion and can pass through the cells lining the lung into the interstitial space.

The very high surface areas of the ultrafine particles can result in a faster release of toxic compounds than would occur from larger particles of the same composition.

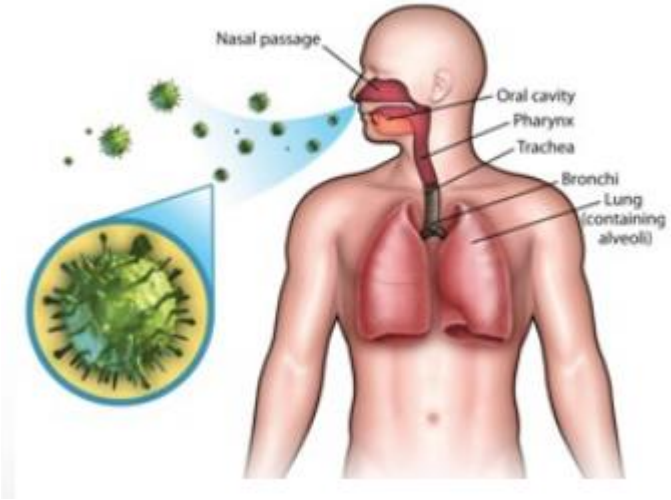
Human Health effects (2)

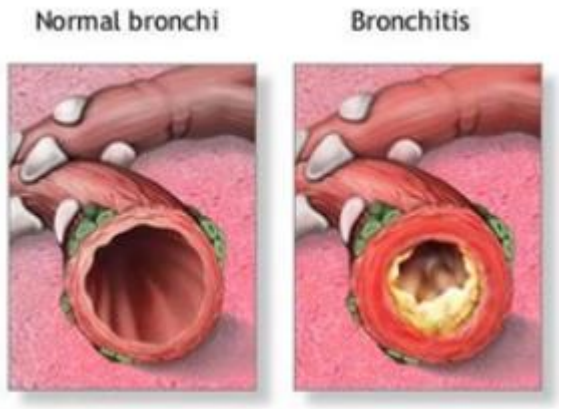


The radioactivity recorded over the liver and bladder expressed as a percentage of the initial lung radioactivity.
(from F. Forestiere, **Health effects of air pollution**, Lincei 2005.)

Human Health effects (3)

In the U.S. half of all lungs examined at autopsy show alveolar deterioration.
Smoking is the most important air pollution in the U.S.
400,000 die annually from diseases related to smoking. This is 20% of all mortality.
Associated costs are estimated at $100 billion annually.





Aerosols and water

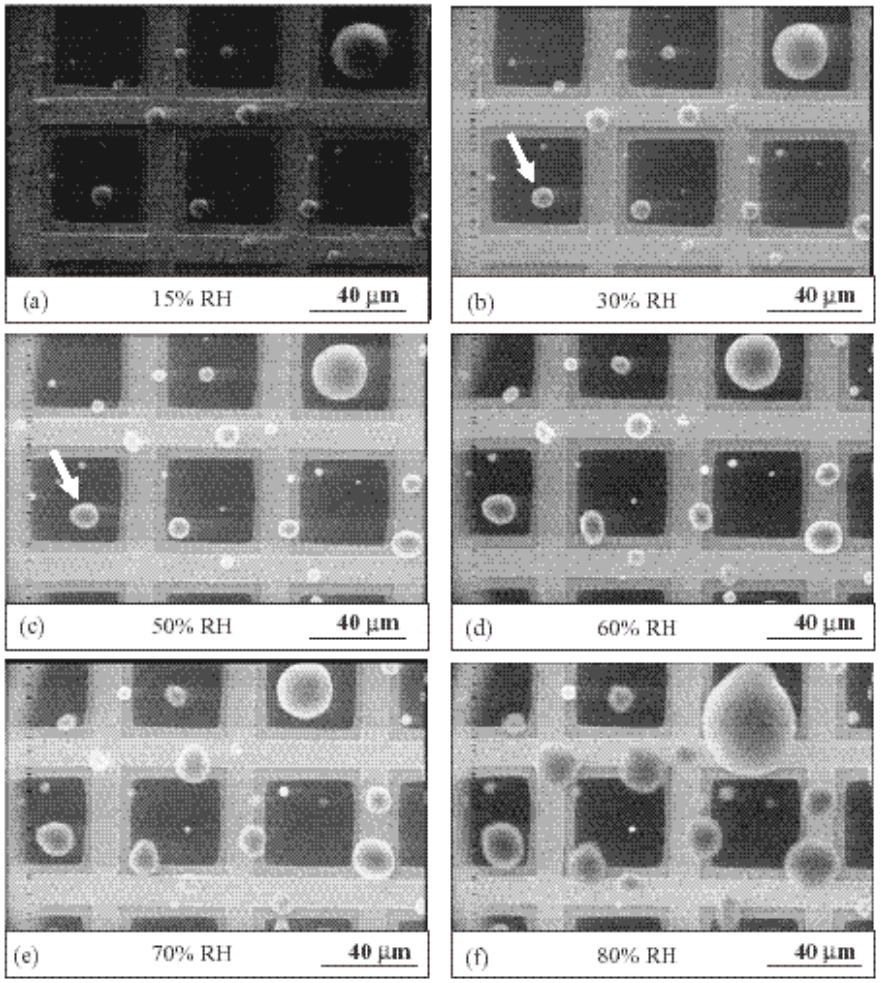
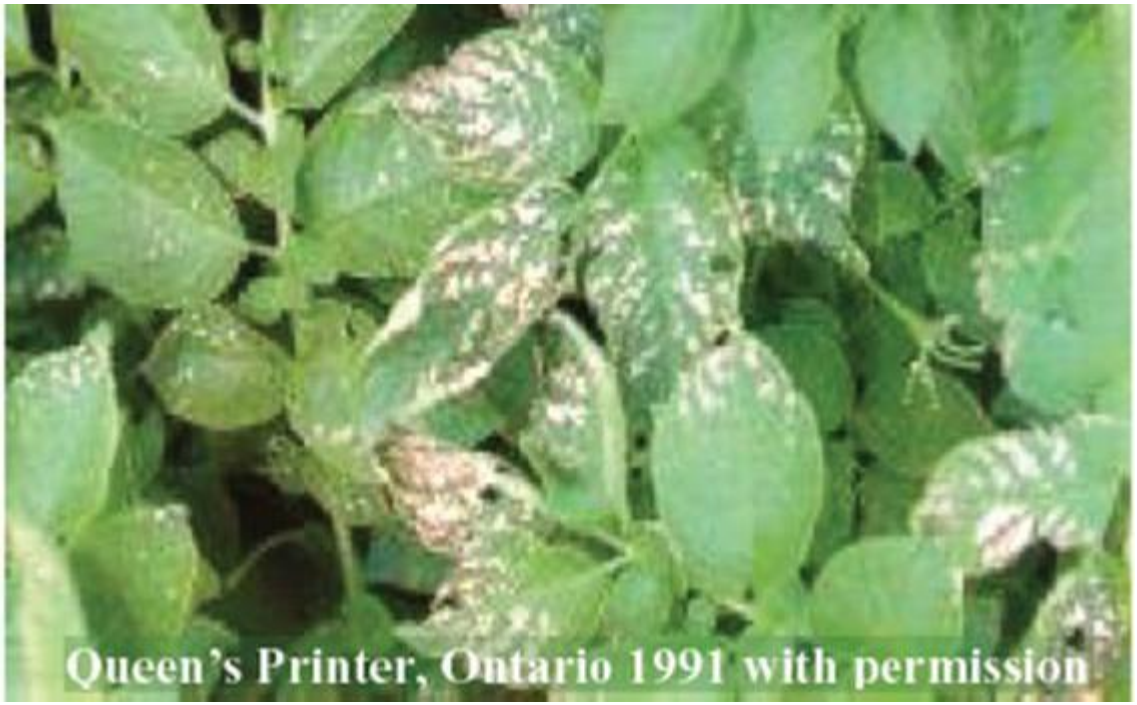


Fig. 2. ESEM images of $NaNO_3$ aerosol particles growing in size upon increasing water vapor pressure: (a) 15% RH, (b) 30% RH, (c) 50% RH, (d) 60% RH, (e) 70% RH and (f) 80% RH. The arrow points at an example of a particle that has increased in size between 30% and 50% RH.

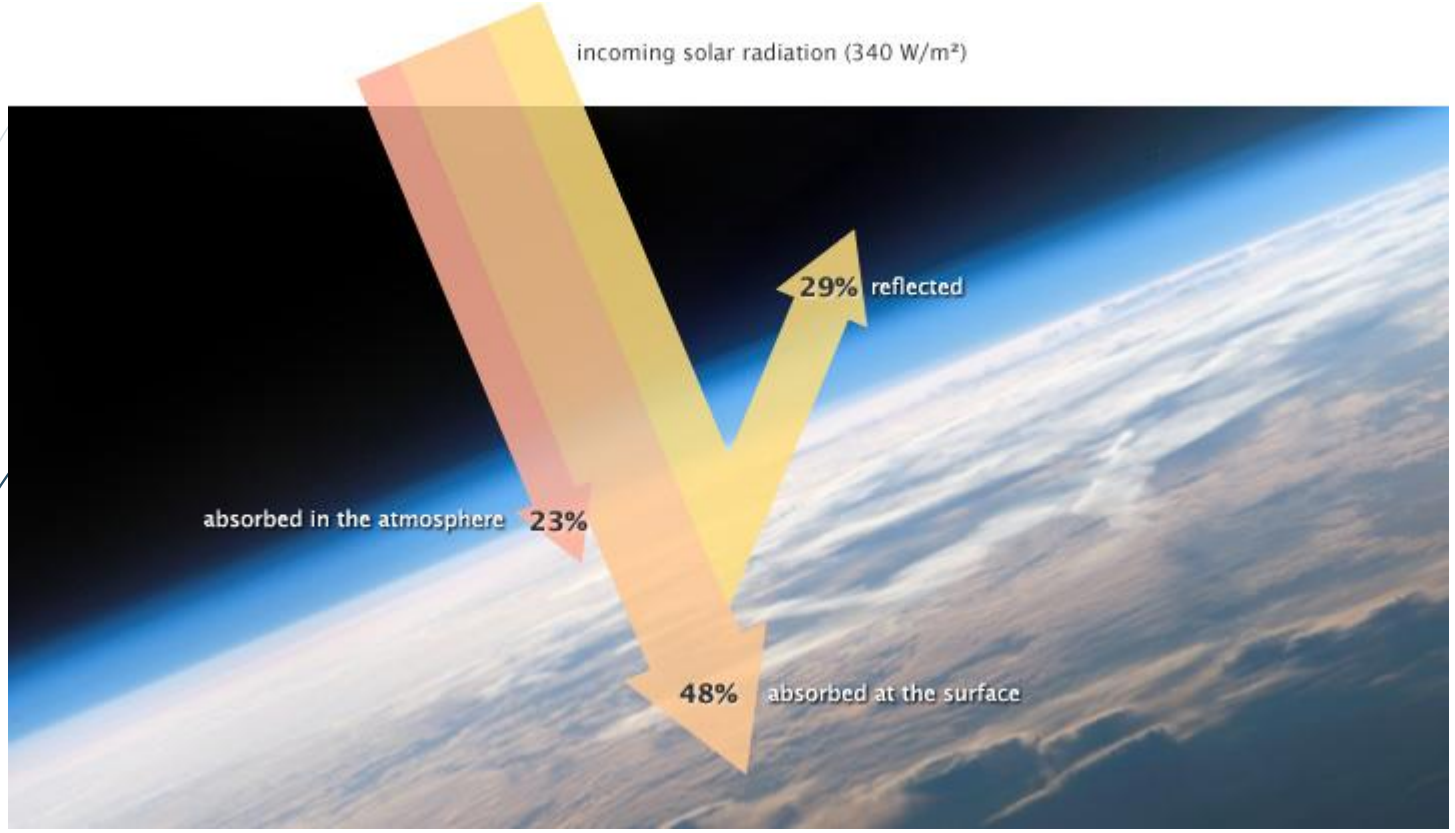
Aerosols and plants

Chemical pollutants can directly damage plants or can cause indirect damage by reducing yields.



Potato leaves damaged by sulfuric acid.

RADIATIVE FORCING



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RADIATIVE FORCING

"Radiative forcing is a measure of the influence a factor has in altering the balance of incoming and outgoing energy in the Earth-atmosphere system and is an index of the importance of the factor as a potential climate change mechanism. In this report radiative forcing values are for changes relative to preindustrial conditions defined at 1750 and are expressed in Watts per square meter (W/m^2)."

Intergovernmental Panel on Climate Change (IPCC) AR4 report

Radiative forcing (1)

"Radiative forcing was very small in the past, when global average temperatures were not rising or falling substantially."
Ronald Prinn, the TEPCO Professor of Atmospheric Science and director of MIT's Center for Global Change Science

For convenience, most researchers choose a "baseline" (1750 or 1850) year before the beginning of world industrialization as the zero point, and compute radiative forcing in relation to that base.

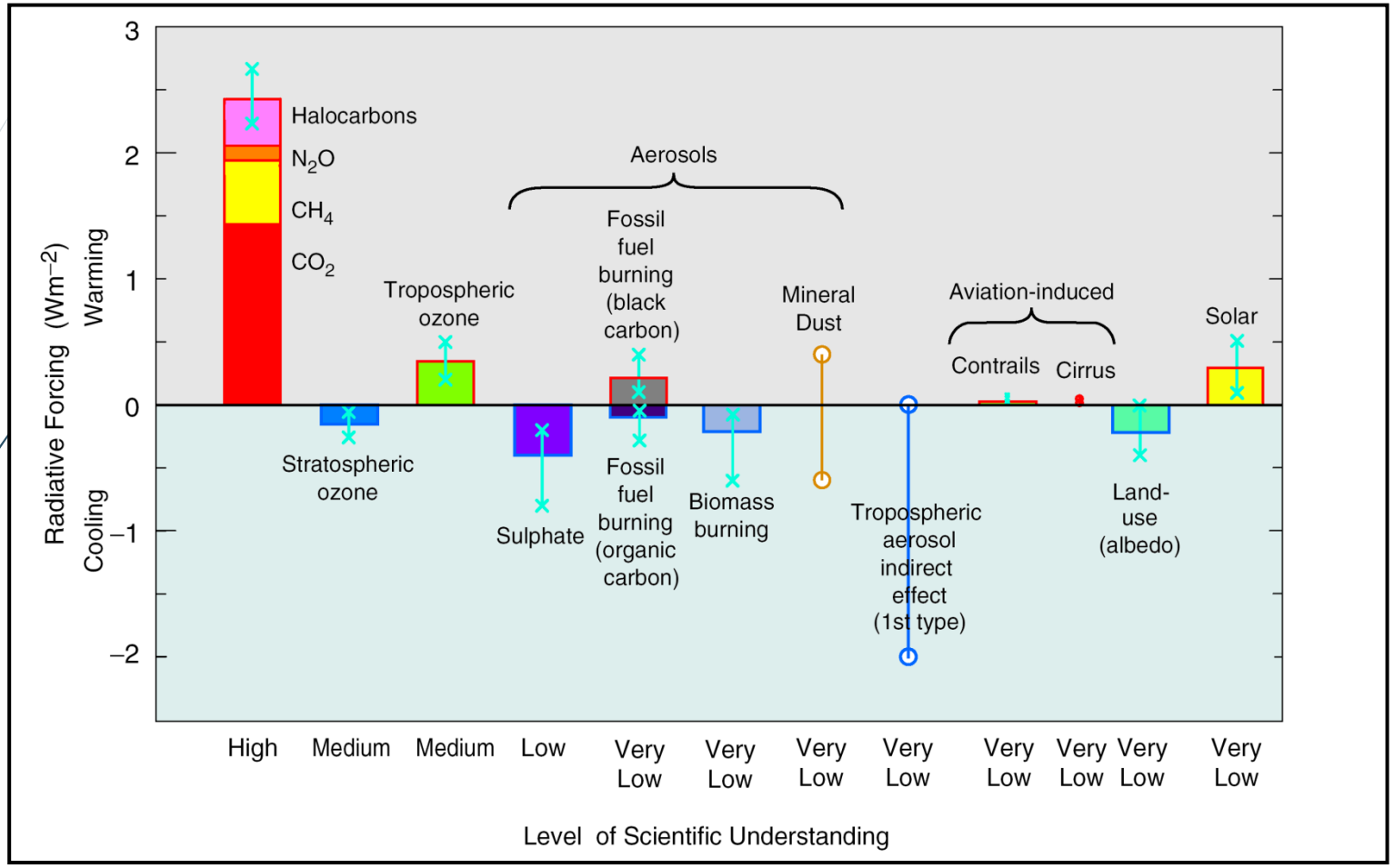
The IPCC uses **1750** as its base year and it is the changes in the various radiative forcing agents since then that are counted.

Radiative forcing (2)

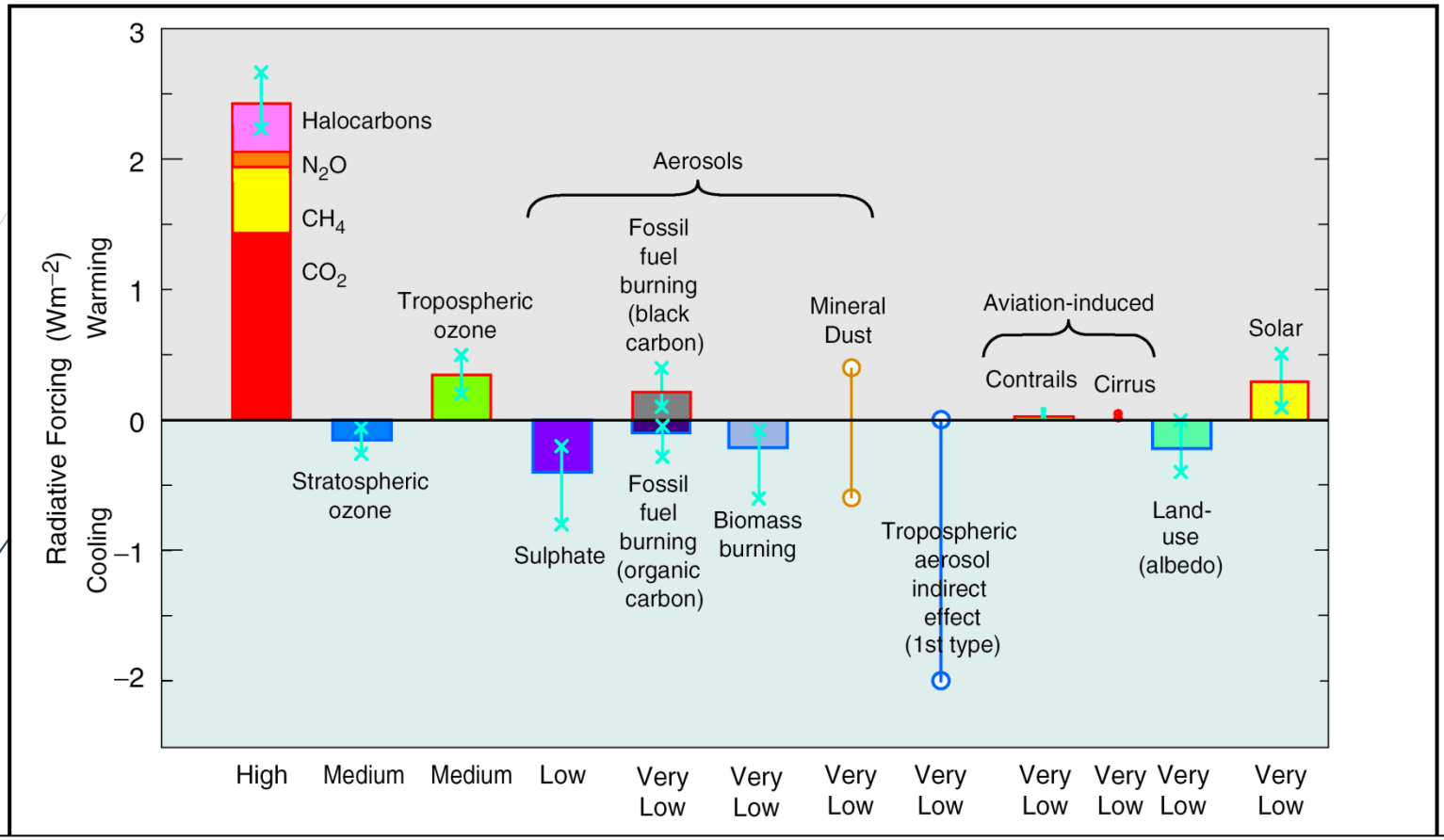
Radiative forcing, measured in watts per square meter of surface, is a direct measure of the impact that recent human activities — including not just greenhouse gases added to the air, but also the impact of deforestation, which changes the reflectivity of the surface — are having on changing the planet's climate.

However, this number also includes any natural effects that may also have changed during that time, such as changes in the sun's output (which has produced a slight warming effect) and particles spewed into the atmosphere from volcanoes.

Radiative forcing (3)

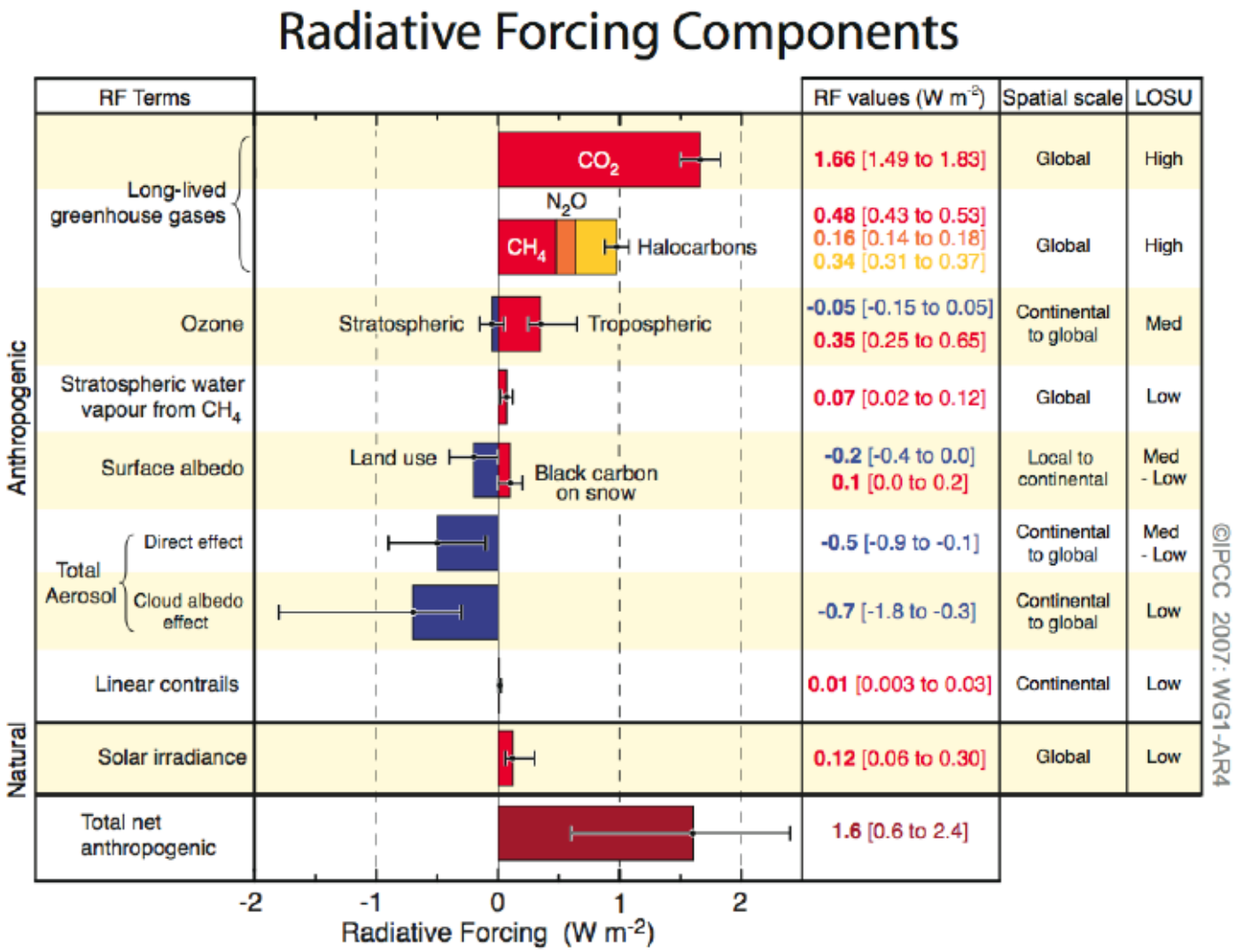


Radiative forcing (4)



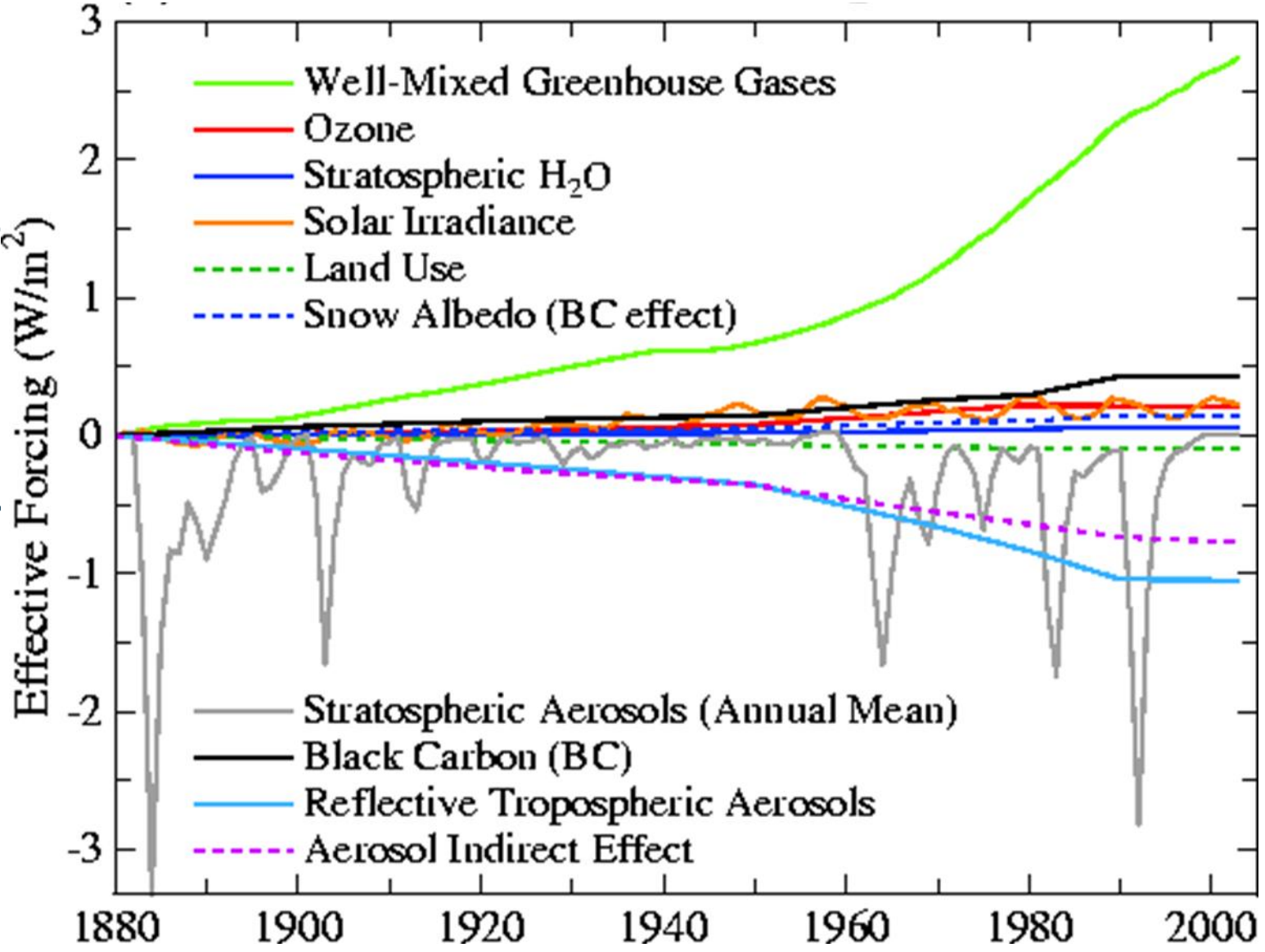
The effects of aerosols are highly complex and often contradictory.
For example, **bright aerosols** (like sulfates from coal-burning) are a cooling mechanism, whereas **dark aerosols** (like black carbon from diesel exhausts) lead to warming.
Also, adding sulfate aerosols to clouds leads to smaller but more abundant droplets that increase cloud reflectivity, thus cooling the planet.

Radiative forcing (5)



The current level of radiative forcing, according to the IPCC AR4, is 1.6 watts per square meter (with a range of uncertainty from 0.6 to 2.4).

Radiative forcing (6)



STRATOSPHERIC AEROSOLS

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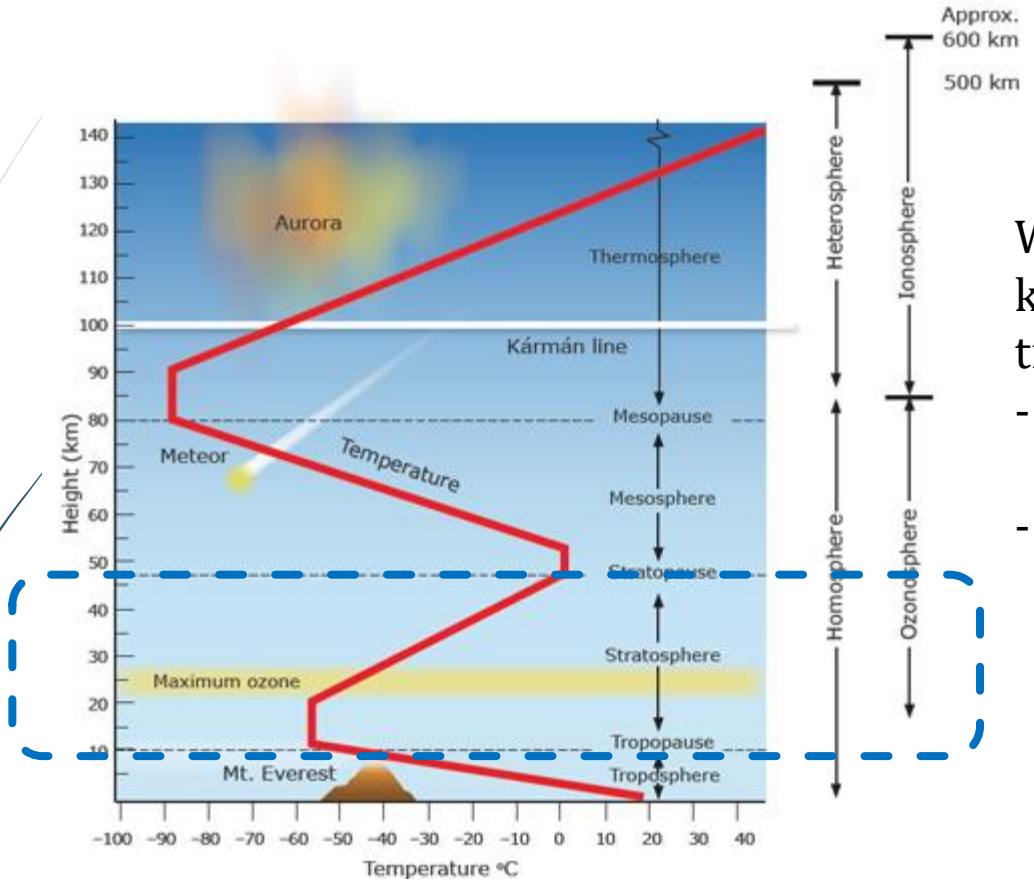
Stratospheric aerosols



Within the first few kilometers above the tropopause:

- **water vapor decreases**
- **O_3 concentrations** often **increase by an order of magnitude**

There is very little vertical mixing between the relatively moist, ozone-poor troposphere and the dry, ozone-rich stratosphere.

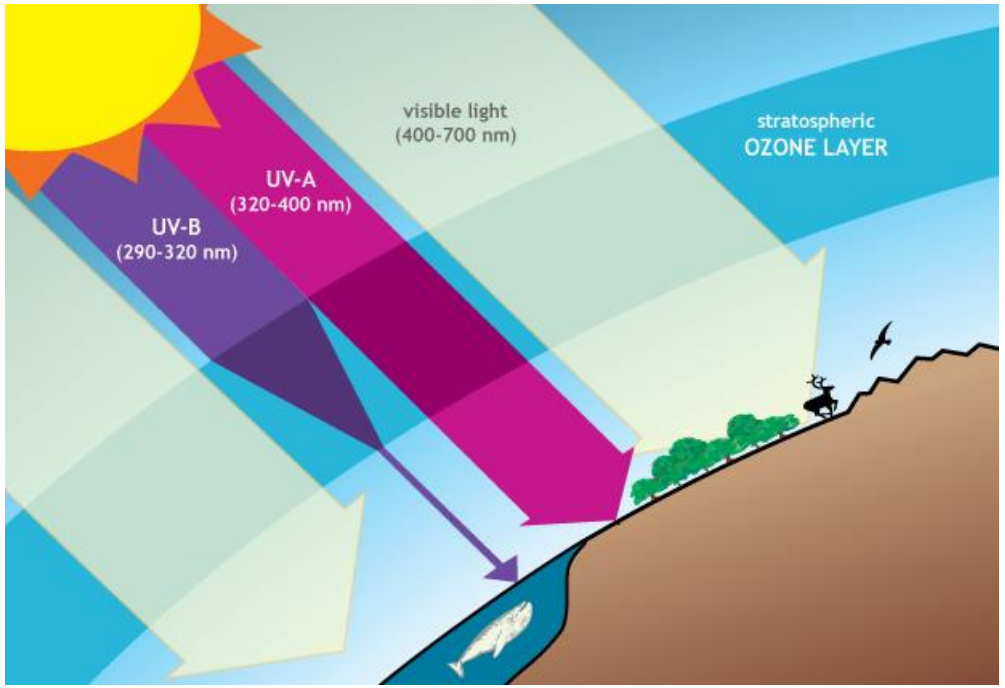
Stratospheric aerosols

Within the stratosphere the air is generally **neutral** or **stable** with respect to vertical motions.

The **removal of aerosols and trace gases by precipitation**, which is a powerful cleansing mechanism in the troposphere, **is generally absent in the stratosphere**.

Materials that enter the stratosphere (e.g., volcanic effluents, anthropogenic chemicals that diffuse across the tropopause or are carried across the tropopause by strong updrafts in deep thunderstorms, and effluents from aircraft) **can remain there for long periods of time**, often as stratified layers.

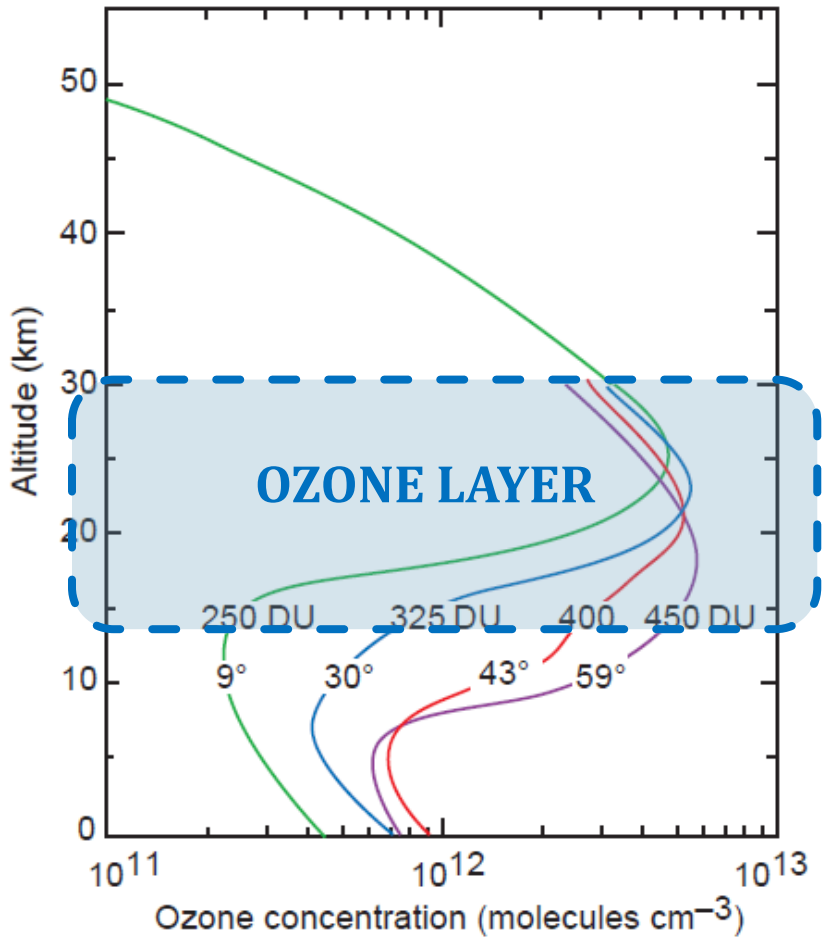
Unperturbed Stratospheric Ozone



- Forms a protective shield that reduces the intensity of UV radiation (with wavelengths between 0.23 and 0.32 μm) from the sun.
- It determines the vertical profile of temperature in the stratosphere because of the absorption of UV radiation.
- It is involved in many stratospheric chemical reactions

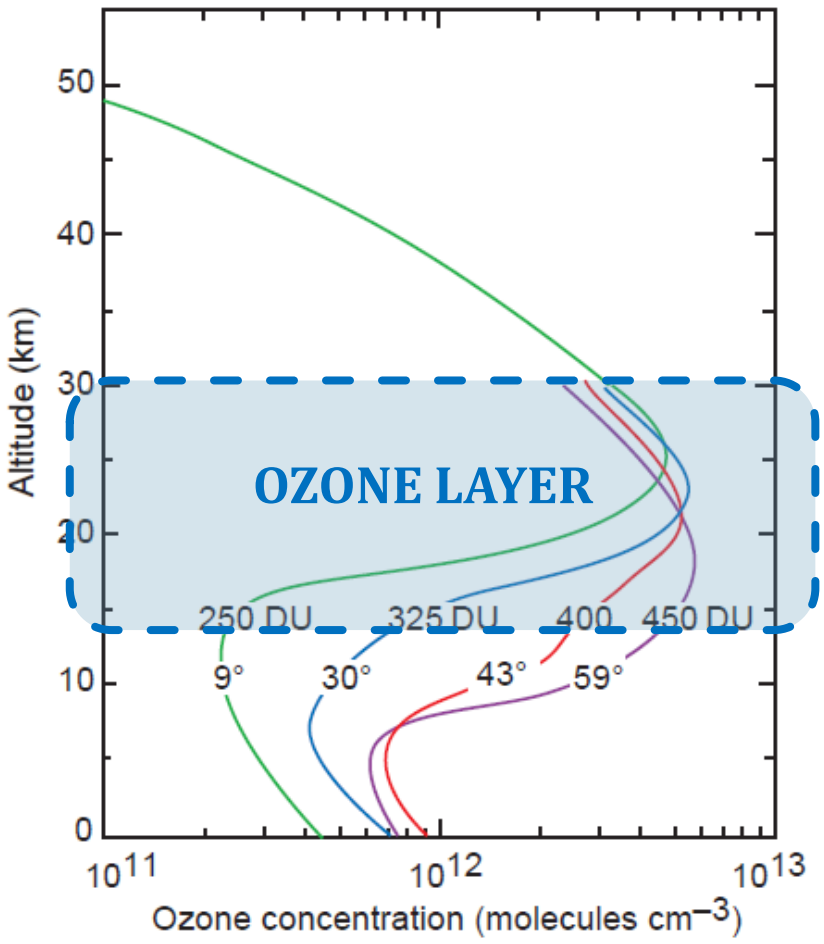
Unperturbed Stratospheric Ozone

Mean vertical distributions of ozone concentrations based on measurements at different latitudes.



Its height and intensity change with latitude, season, and meteorological conditions.

Unperturbed Stratospheric Ozone

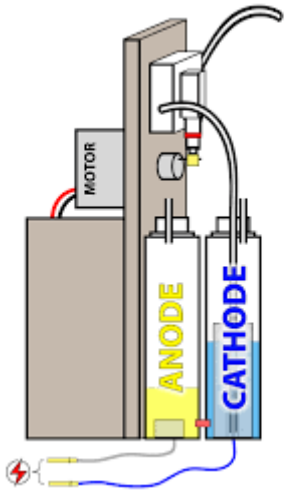


- greatest column densities of O_3 in the northern hemisphere occur in polar latitudes in spring; in the southern hemisphere the spring maximum is at midlatitudes
- maximum production over the tropics (photochemical reactions)
- peaks in concentrations at polar and midlatitudes are attributed to meridional and downward transport of O_3 away from the equator

Remote sensing measurements of O_3 (1)

Vertical profile of O_3 via ozonesonde:

- Two electrolytic cells, containing a solution of potassium iodide, are initially in chemical and electrical equilibrium.
- When an air sample containing O_3 is drawn through one of the cells, the equilibrium is perturbed and an electric current flows between the cells.
- The amount of electric charge, proportional to the partial pressure of the O_3 in the ambient air, is continuously transmitted to a ground station.



Remote sensing measurements of O_3 (2)

Column O_3 via Dobson spectrophotometer:

Measurement of the amount of UV sunlight that reaches the ground, deducing from this how much UV absorption occurred due to O_3.

Absorption by O_3 occurs in the UV-B region (λ=0.290–0.320 μm).

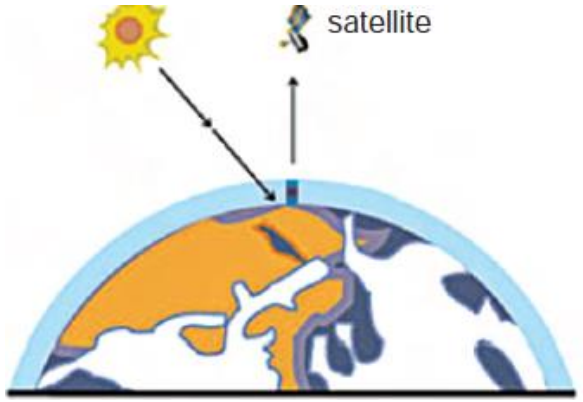
However, clouds and some aerosol particles also absorb in this wavelength band.

By rationing the two measured values, absorption by O_3 in the total vertical column can be obtained.

Remote sensing measurements of O_3 (3)

Column O_3 via satellite:

Ozone can be derived from satellite observations four passive techniques.



1) Backscattering UV (BUV):

Two pairs of measurements are needed: the incoming UV irradiance and the backscattered UV radiance at two wavelengths: the former strongly absorbed by O_3 and the latter weakly absorbed by O3.

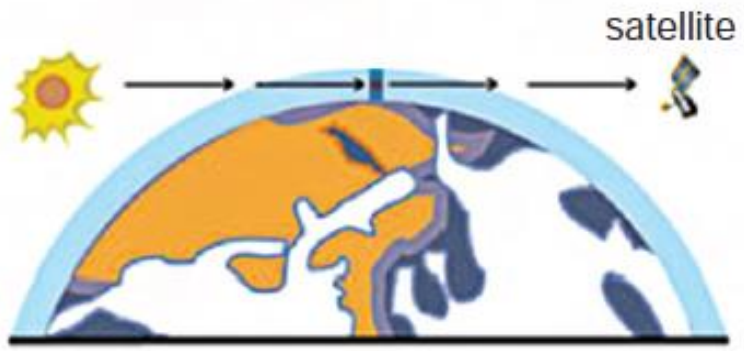
The difference between these two pairs of measurements can be used to infer the total O_3 column.

Vertical profiles of O_3 can be obtained by measuring backscattered radiation at a number of wavelengths.

Remote sensing measurements of O_3 (4)

Column O_3 via satellite:

Ozone can be derived from satellite observations four passive techniques.



2) Occultation

This technique has been used to monitor O3 since 1984.

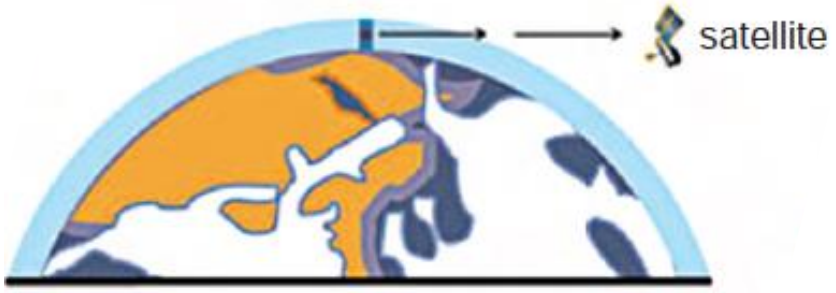
Measure radiation through the limb of the atmosphere when the sun, moon, or a star is rising or setting.

From measurements of the amounts of radiation at various wavelengths absorbed by the atmosphere, vertical profiles of various trace constituents can be derived.

Remote sensing measurements of O_3 (5)

Column O_3 via satellite:

Ozone can be derived from satellite observations four passive techniques.



3) Limb emission

Measurements of infrared or microwave radiation emitted by the atmosphere along the line of sight of the instrument.

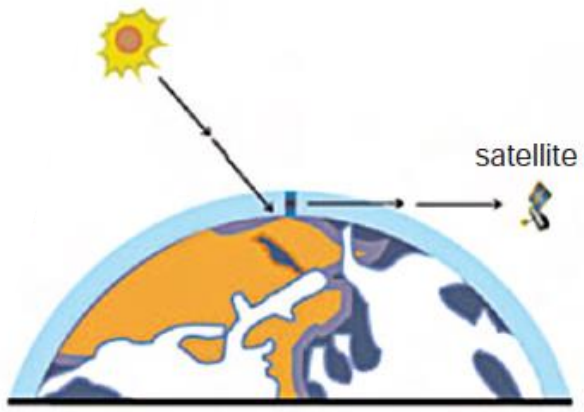
Microwave emission passes through clouds and can provide measurements lower in the atmosphere than infrared emissions.

However, microwave detectors are large, heavy, and require more power than infrared detectors.

Remote sensing measurements of O_3 (6)

Column O_3 via satellite:

Ozone can be derived from satellite observations four passive techniques.



4) Limb scattering

Measures scattered light rather than light directly from the sun or moon. Consequently, if the sun is visible, this technique can provide essentially continuous measurements. Limb scattering works best with O_3, but H_2O(g), NO_2, SO_2, and aerosols can also be measured.

The Ozone Hole (1)

- Chlorofluorocarbons (CFCs) were first synthesized in **1928** as the result of a search for a nontoxic, nonflammable refrigerant.
  They were marketed under the trade name Freon and became widely used as refrigerants, propellants in aerosol cans, inflating agents in foam materials, solvents and cleansing agents.

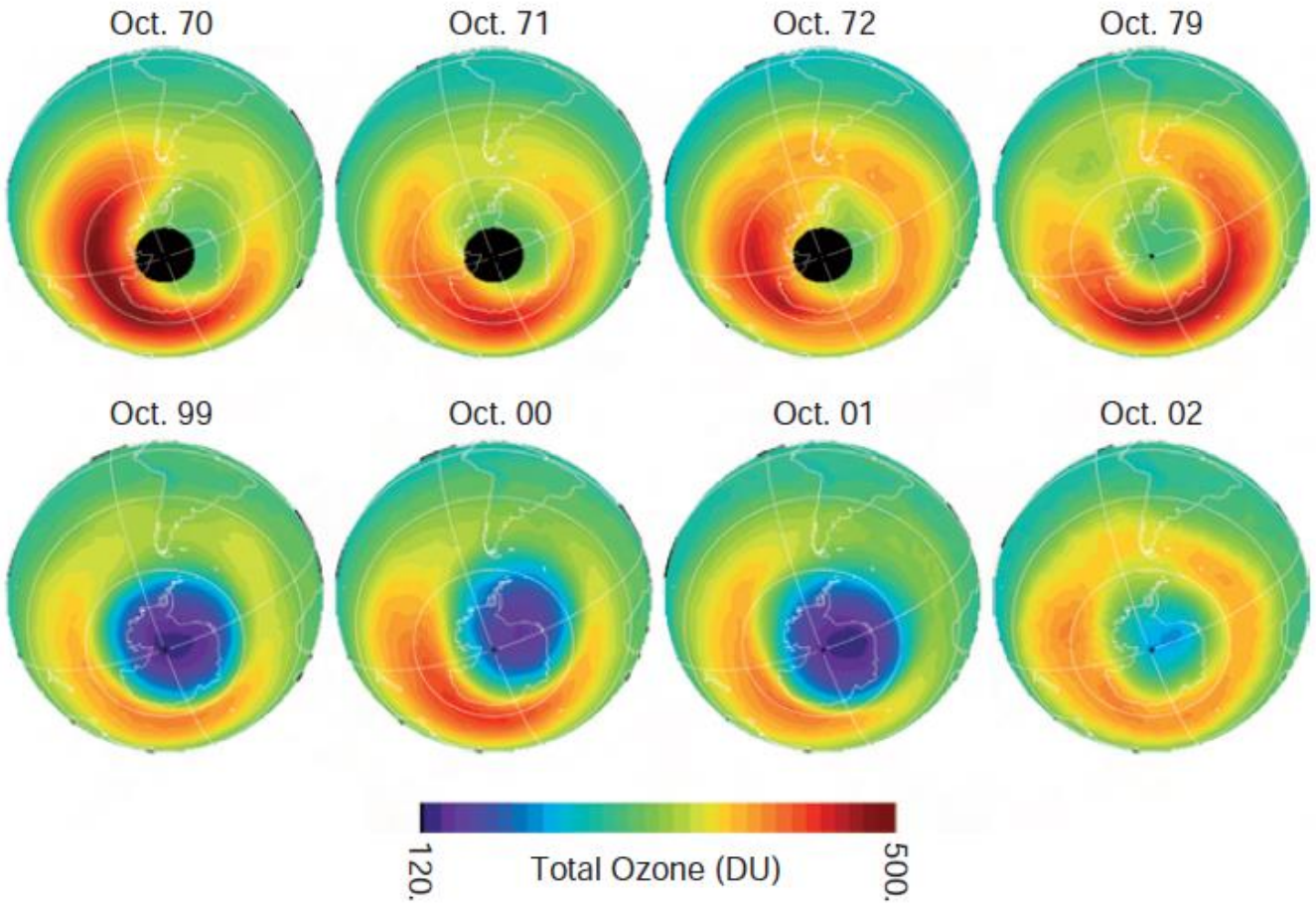
- In **1973** it was found that CFCs were spreading globally and, because of their inertness, were expected to have residence times ranging up to several hundred years in the troposphere

  Such long-lived compounds eventually find their way into the stratosphere where, at altitudes of 20 km, they absorb UV radiation in the wavelength interval 0.19–0.22 μm and photodissociate.
  Because CFCs absorb strongly in the infrared, they are also significant greenhouse gases.

- In **1985**, British scientists, carried out for many years remote sensing measurements of O_3 at Halley Bay in the Antarctic, reported a 30% decrease in springtime total column O_3 since 1977.

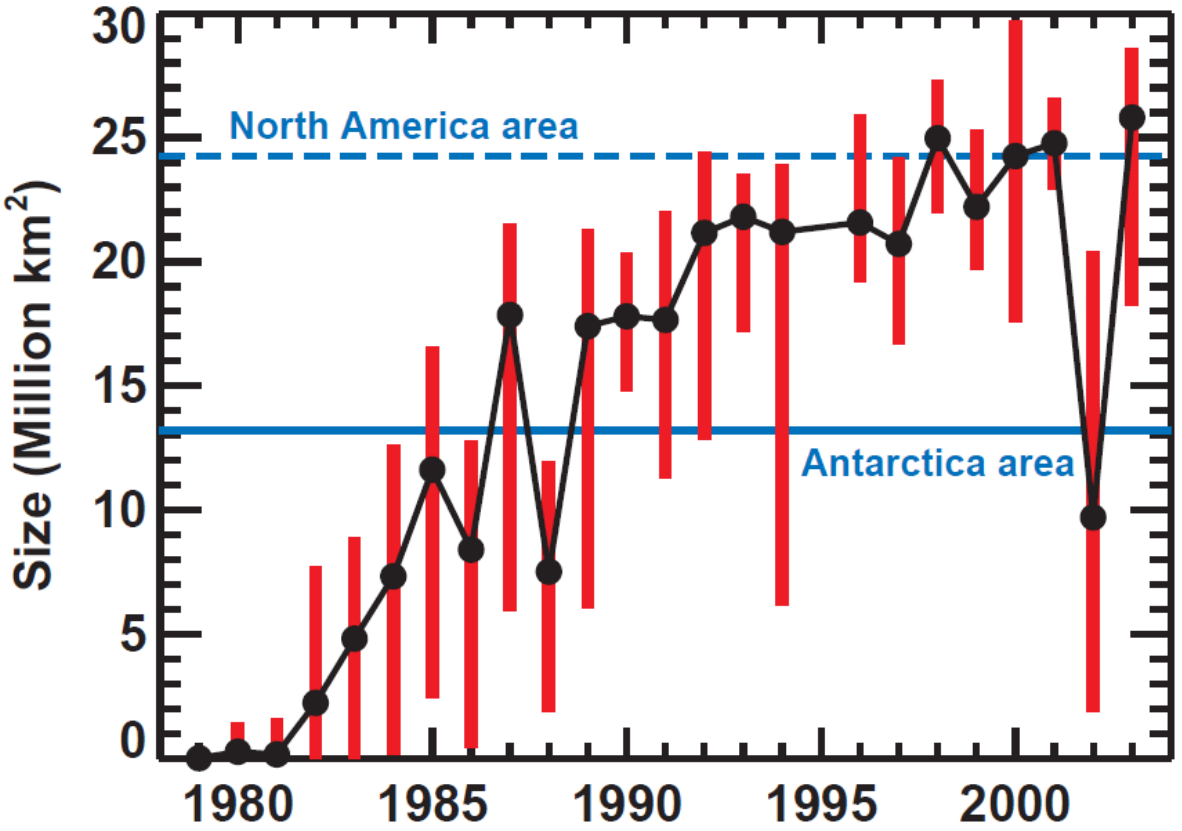
  These observations were subsequently confirmed by remote sensing measurements from satellite and by airborne in situ measurements.

The Ozone Hole (2)



Anomalously low O_3 values (purple) over the continent in October from 1999 to 2002.

The Ozone Hole (3)

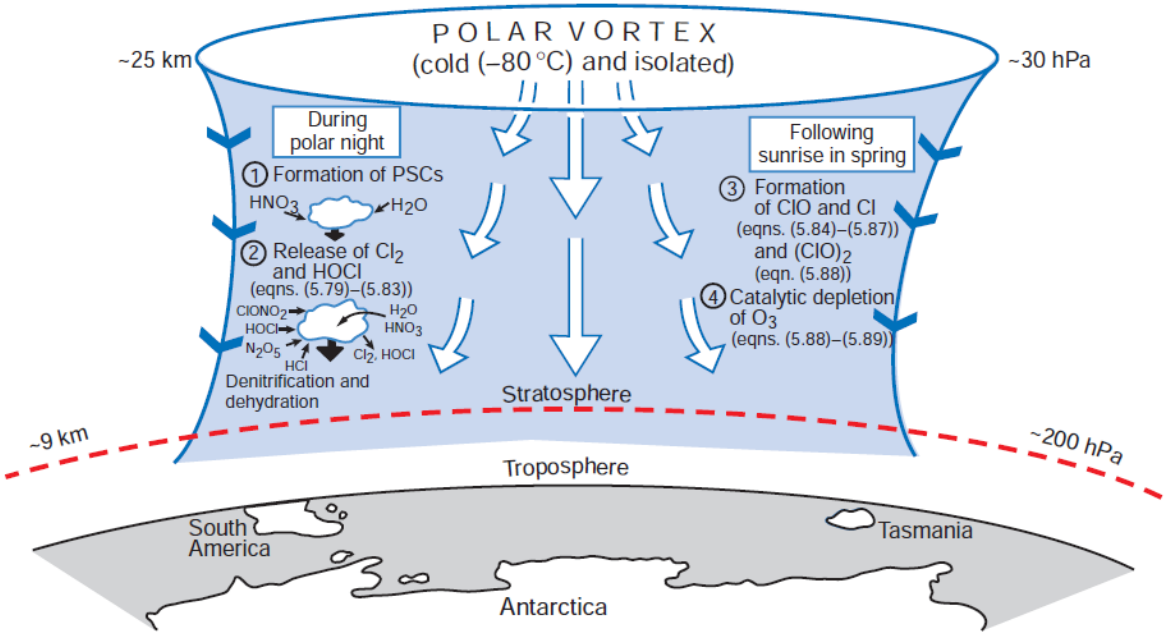


Year-to-year variations are caused by temperature fluctuations near the polar vortex edge.
Warmer years have smaller holes.

The Ozone Hole (4)

- Why over the Antarctic?
- Why during spring?
- Why was the size of the ozone hole so much less in 2002 than in other recent years?
- Does an ozone hole, similar to that in the Antarctic, develop in the Arctic stratosphere?

The Ozone Hole (5)

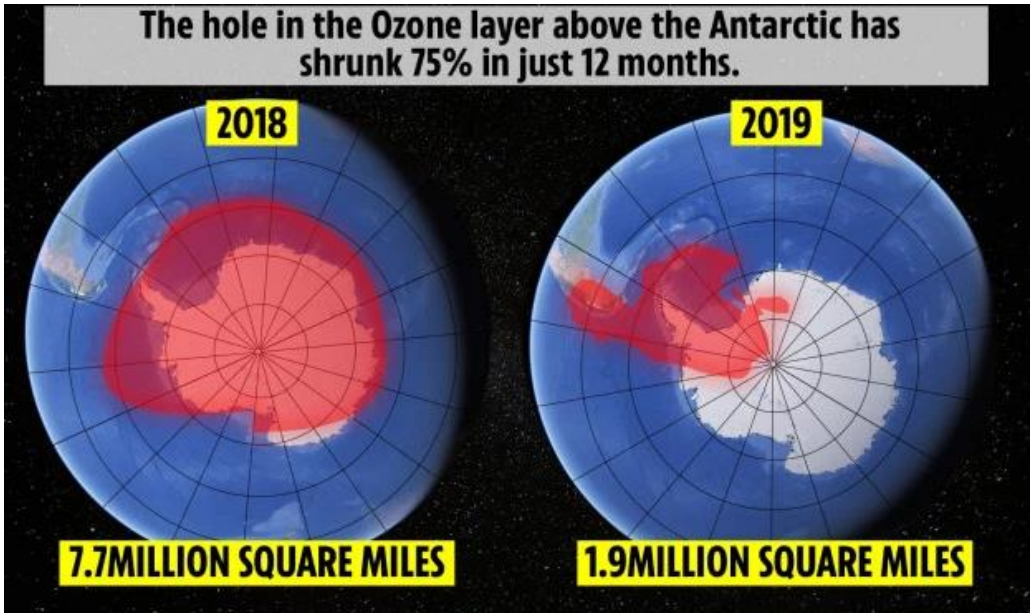


During the austral winter, stratospheric air over the Antarctic continent is restricted from interacting with air from lower latitudes by a large-scale vortex circulation, which is bounded at its perimeter (called the *vortex collar*) by strong westerly winds encircling the pole.

Because of the lack of solar heating in the austral winter, the air within the vortex is extremely cold. High-level clouds, called *polar stratospheric clouds (PSCs)*, form in the cold core of the vortex, where temperatures can fall below -80 °C.

In the austral spring, as temperatures rise, the winds around the vortex weaken and the vortex disappears. However, during winter, the vortex serves as a giant and relatively isolated chemical reactor in which unique chemistry can occur in order to balance the natural content of O_3.

The Ozone Hole (6)



In 2019, the hole that developed in the ozone layer over Antarctica was the smallest on record since the ozone hole was first discovered, according to scientists at NASA and the US National Oceanic and Atmospheric Administration.

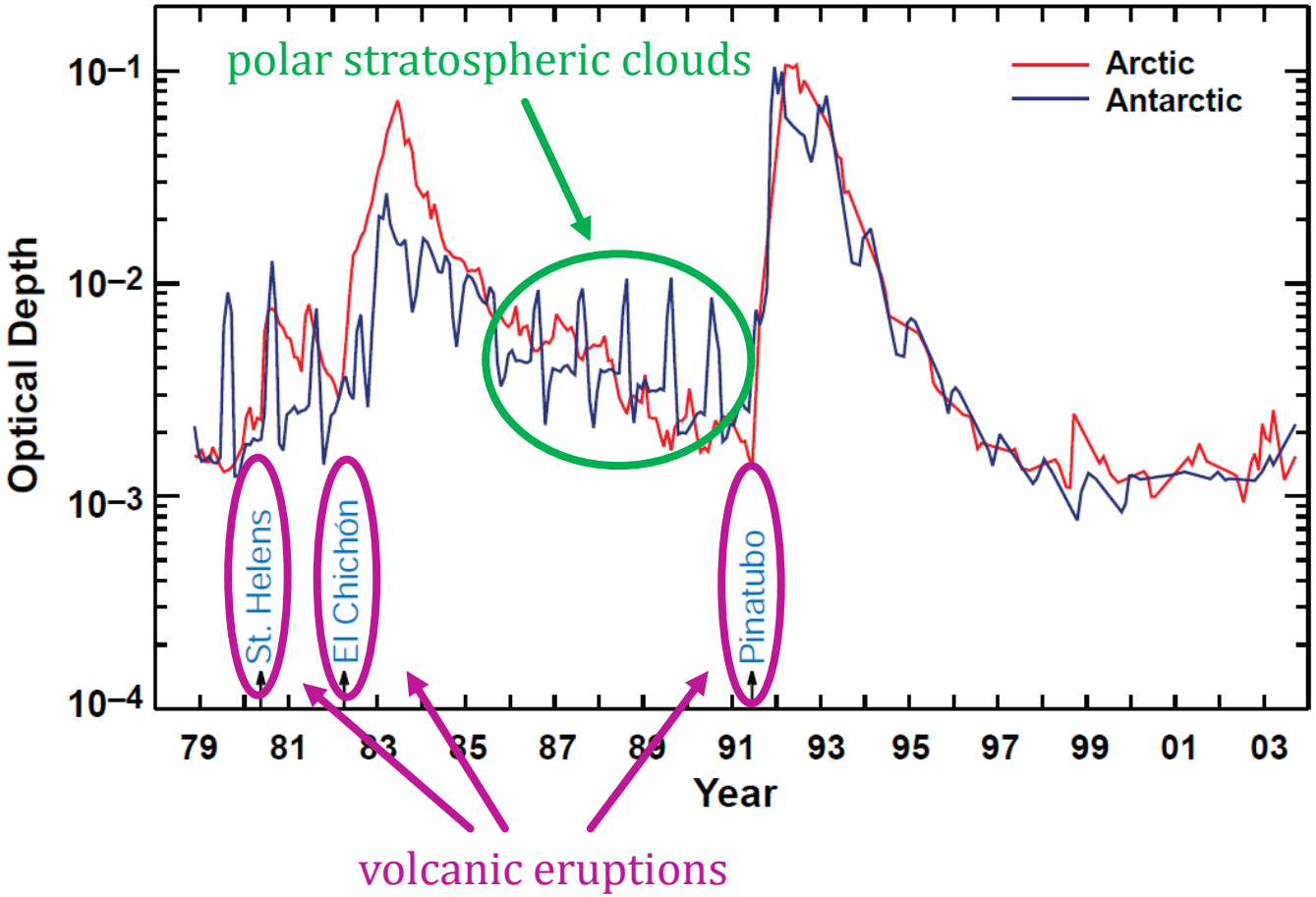
"It's great news for ozone in the Southern Hemisphere but it's important to recognize that what we're seeing this year is due to warmer stratospheric temperatures. It's not a sign that atmospheric ozone is suddenly on a fast track to recovery."

Paul Newman,
chief scientist for Earth Sciences at NASA's Goddard Space Flight Center in Greenbelt.

Sulfur in the Stratosphere (1)

Particles with radii ≈0.1–2 μm reach a maximum concentration at altitudes of about 17–20 km in the region called the **stratospheric sulfate layer (Junge layer).**

These particles are composed of about 75% sulfuric acid (H_2SO_4) obtained from SO_2 emitted by volcanic eruptions.



Sulfur in the Stratosphere (2)

Eruption of Mt. Pinatubo (Philippines) in 1991.
During the eruption of Mt. Pinatubo, 14-20 Tg (1 Tg = 10^6 tons) of SO_2 was injected into the stratosphere, causing the increase of aerosol concentration from 2-5 μg/m^3 to 20-100 μg/m^3.

URBAN AEROSOLS

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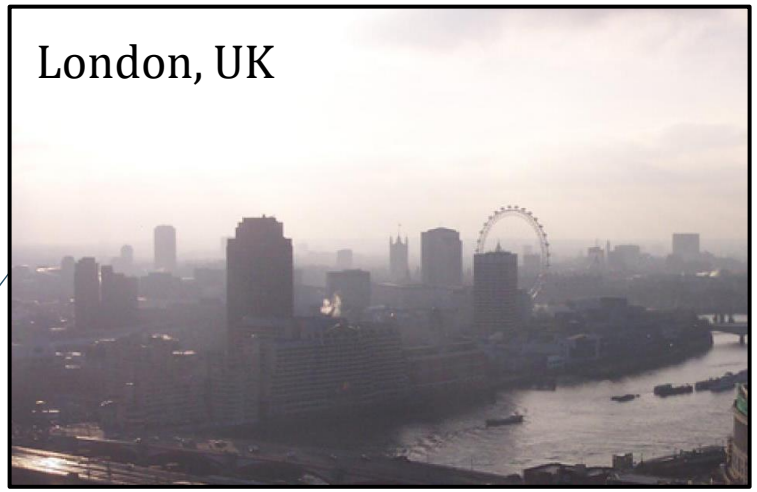
Urban aerosols

- Approximately 147 million tons of air pollutants are released annually into the atmosphere in the U.S.
- Worldwide emissions total around 2 billion metric tons.
- 16 of the 20 smoggiest cities in the world are in China.



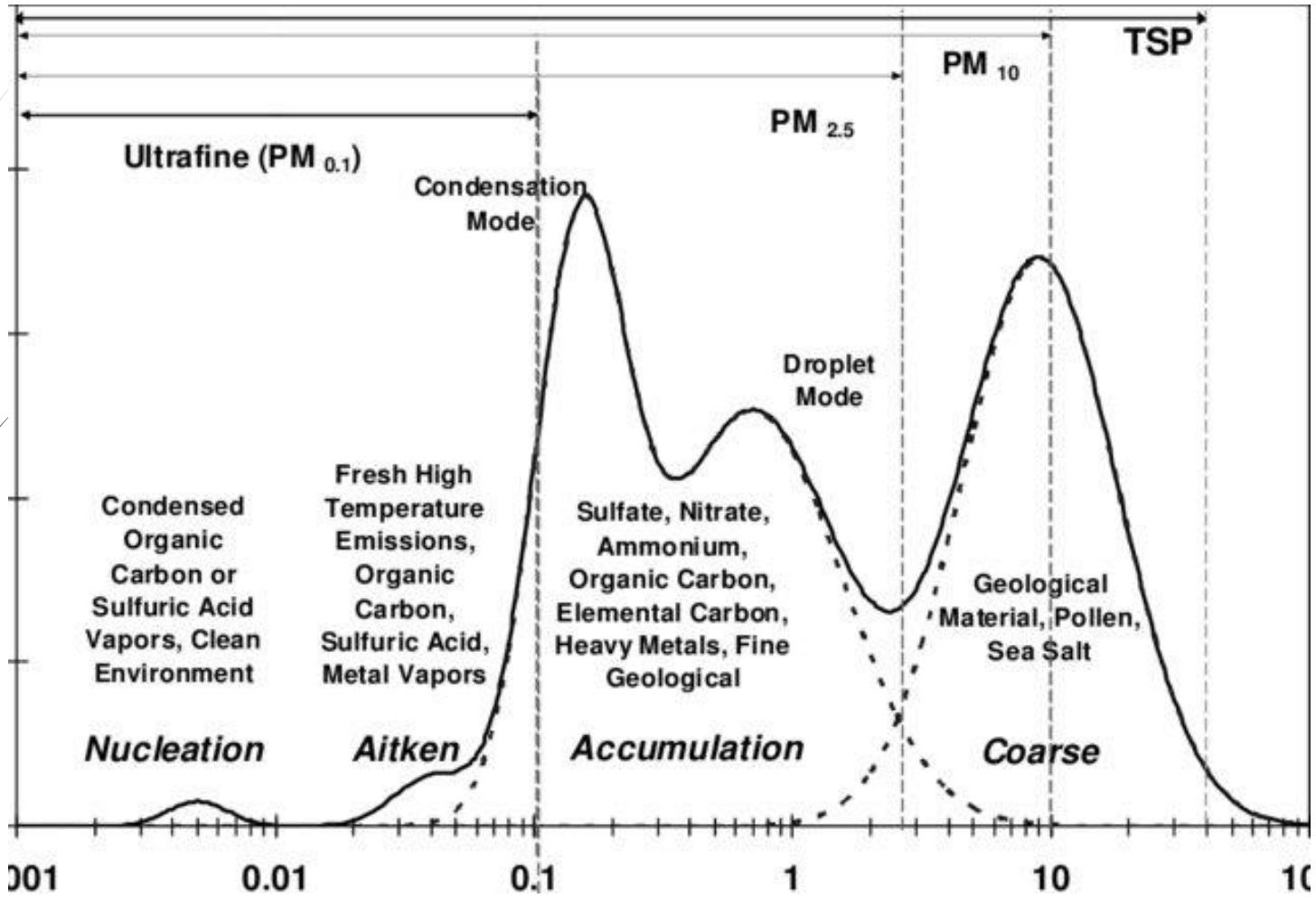
Urban aerosols

- **Anthropogenic sources:**
  - Stationary sources: power plants, mines, refinery plants, etc
  - Traffic
  - Combustion

- **Particle concentration:**
  A few tens of $\mu g/m^3$ to 1 $\mu g/m^3$ in heavily polluted areas

Location	Mass concentration ($\mu g/m^3$)
Background	20
Rural areas	40
Urban areas	
Population $< 10^5$	86
$\approx 10^5 - 10^6$	104
$> 10^6$	154

Urban aerosol size distribution



- Nuclei mode (0.001-0.1 μm)
- Accumulation mode (0.1-2.5 μm)
- Coarse-particle mode (2.5-100 μm)

Urban aerosol size distribution

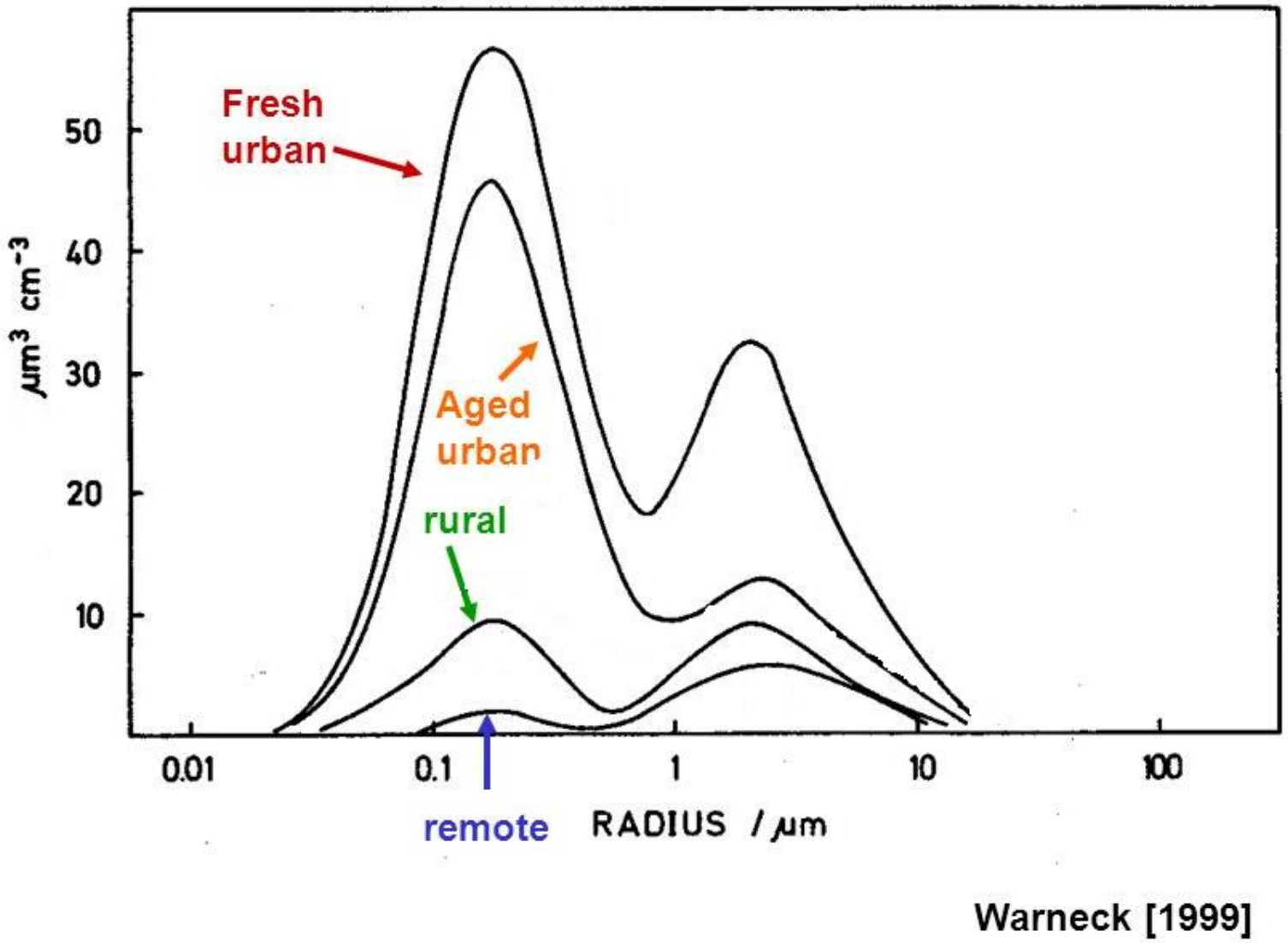
- **Nuclei mode**
  - Size: 0.001-0.1 μm
  - Combustion particles and gas-to-particle conversion
  - Location: near highways and combustion sources
  - High concentration
  - Rapid coagulation

- **Accumulation mode**
  - Size: 0.1-2.5 μm
  - Combustion particles, smog particles, coagulated nulcei mode particles
  - Slow coagulation
  - Account for most of the visibility effects

- **Fine particle**
  - Nuclei + accumulation mode
  - $PM_{2.5}$

- **Coarse-particle mode**
  - Size: 2.5-10 μm
  - Dusts, sea salts, particles from surface mining
  - Ready to settle down on the surface

Urban aerosol size distribution

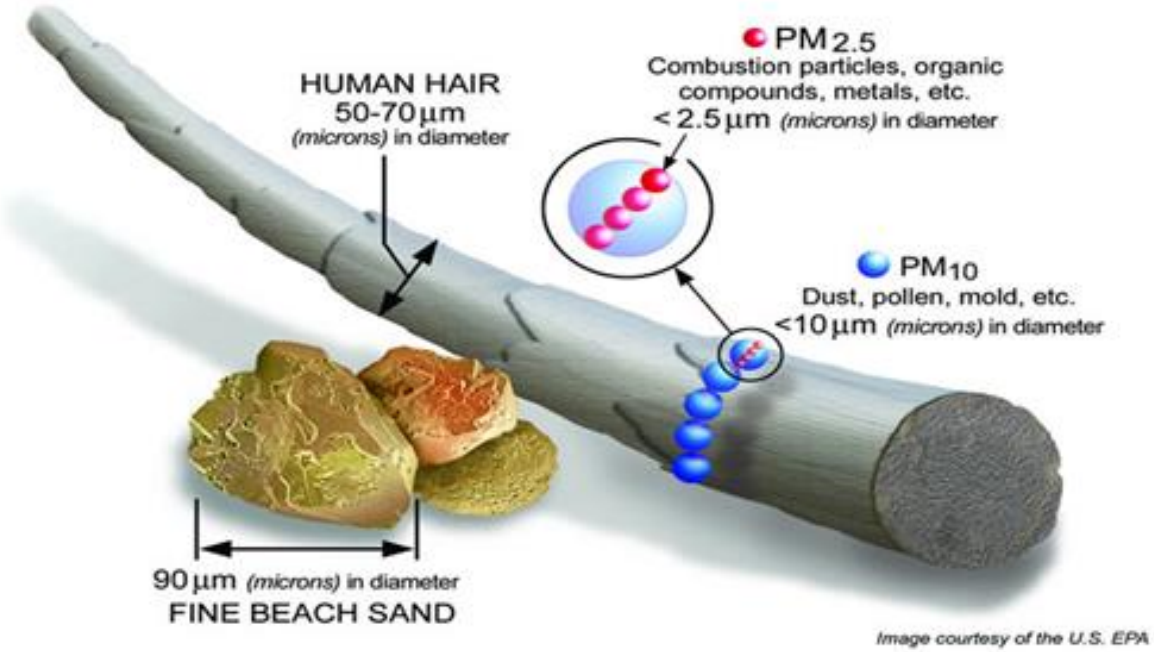


Typical U.S. aerosol size distributions.

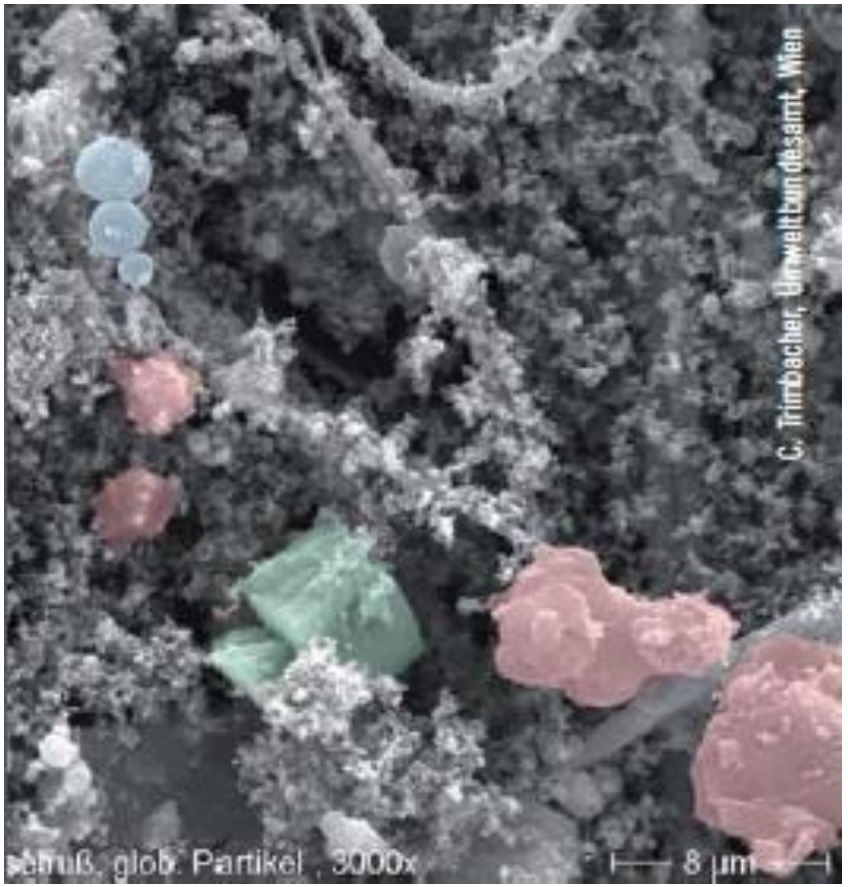
$PM_{2.5}$ and PM_{10} (1)

$PM_{2.5}$ refers to the atmospheric particulate matter that has a diameter of less than 2.5 μm, which is about 3% of the diameter of human hair.

PM_{10} are the particles with a diameter of 10 μm (fine particles), is also known as respirable particulate matter.



$PM_{2.5}$ and PM_{10} (2)



Electron microscopic image of PM on a filter that was placed near a road.

Black carbon (the little, grey balls) is ubiquitous on this filter. The light blue balls are particles originating from combustion processes; while the pink particles are minerals and the green cubes are salts.

Vertical distribution of urban aerosol

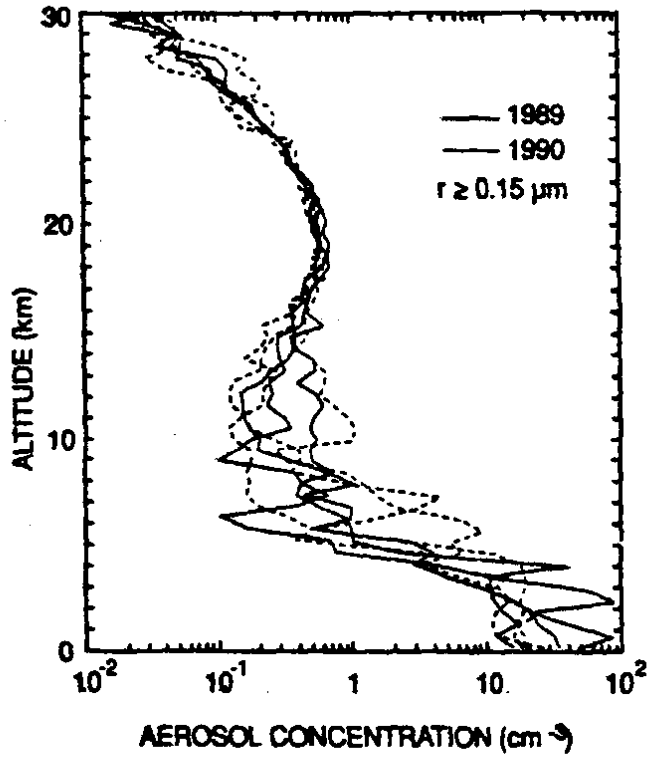


Fig. 8-43: Vertical variation of the concentration of aerosol particles with $r \geq 0.15$ μm in air over Laramie (Wyoming), summer, 1989 and 1990. (From Hofmann, 1993, priv. comm.)

Vertical distribution of urban aerosol

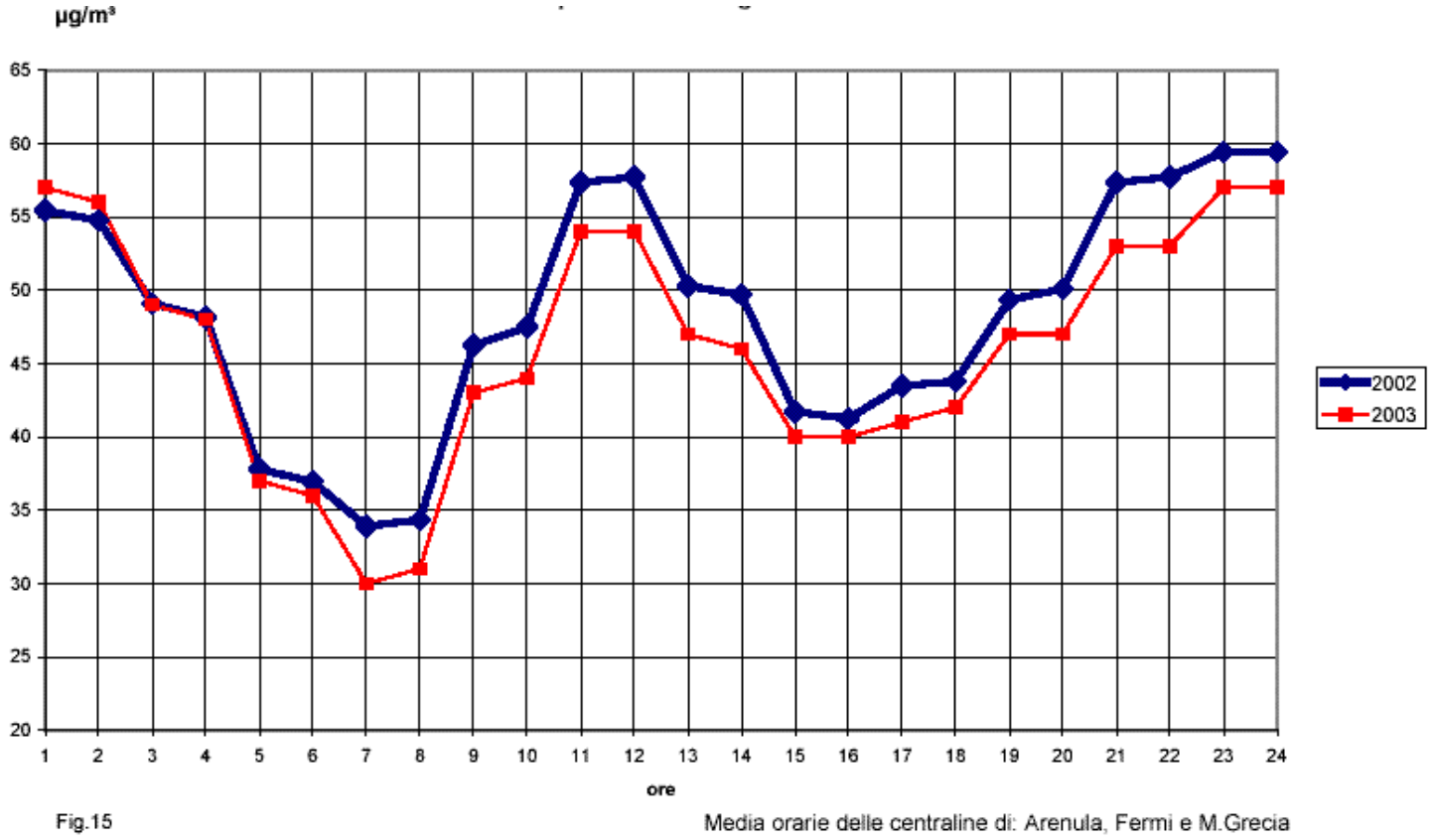
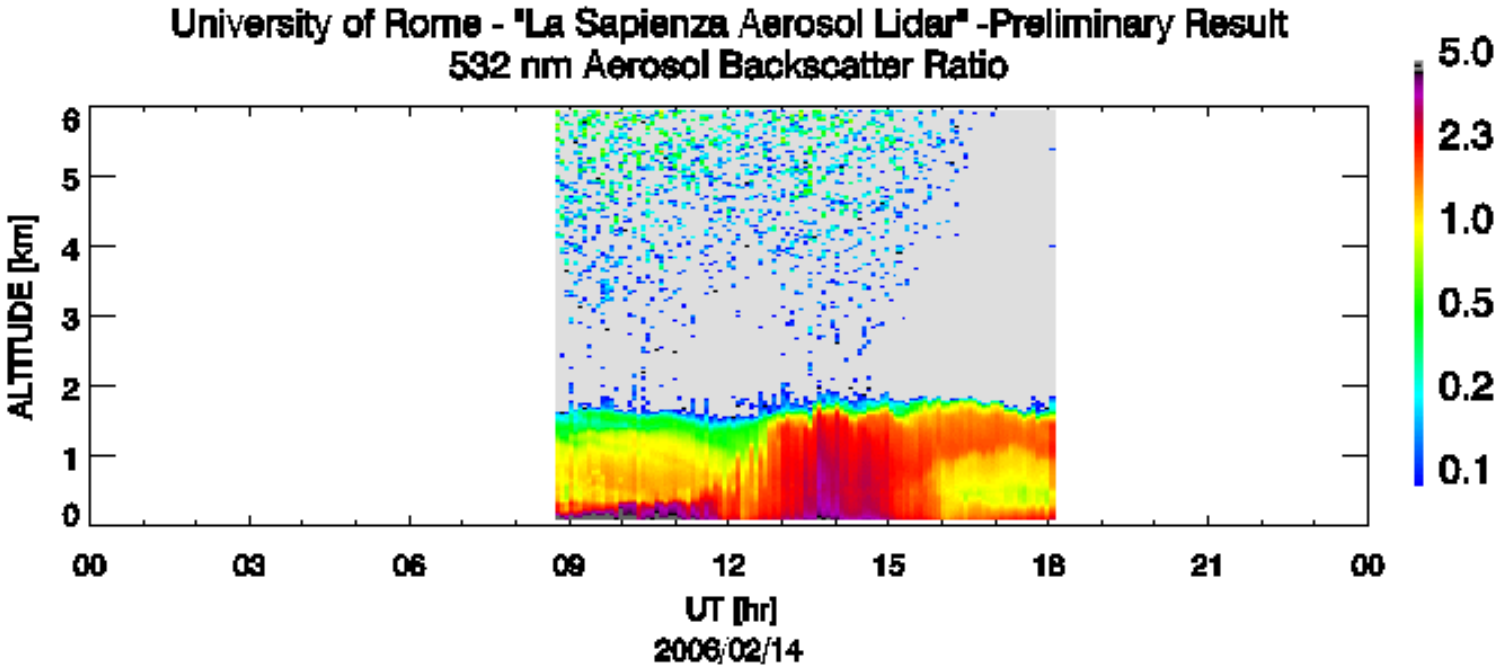


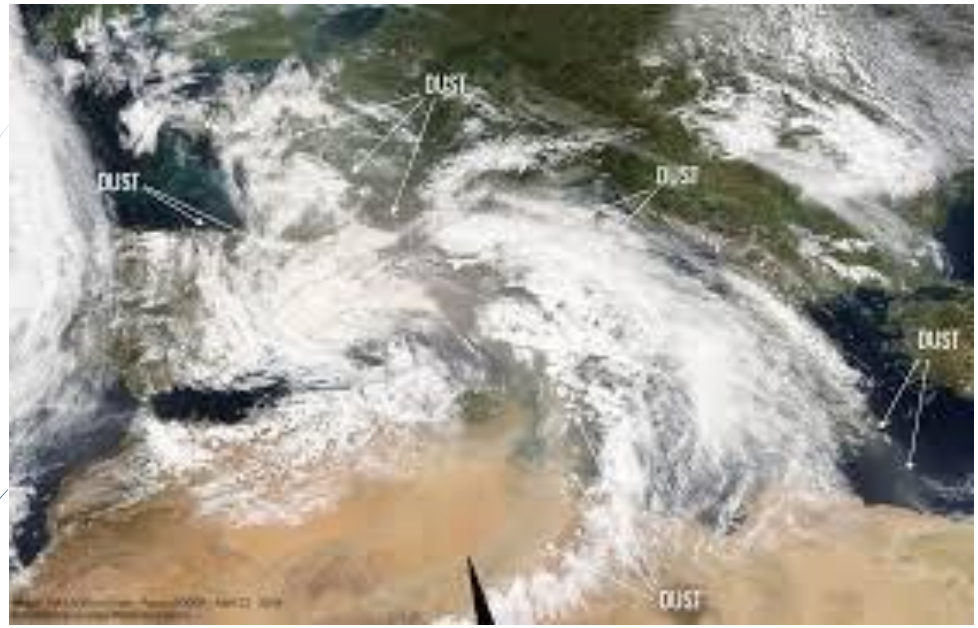
Fig.15 Media orarie delle centraline di: Arenula, Fermi e M.Grecia

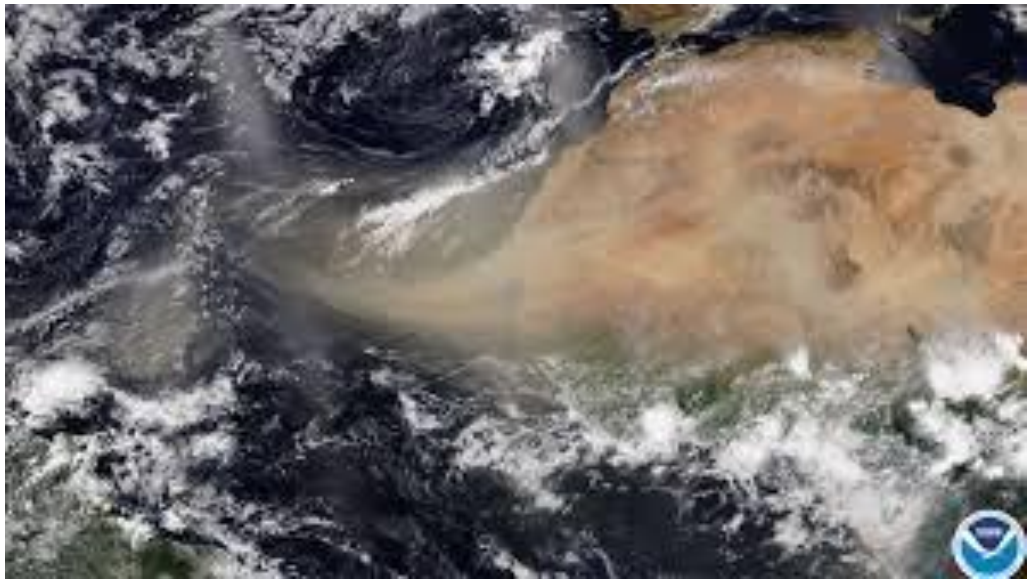
Typical hourly distribution of PM_{10} concentration in 2002-2003.

Vertical distribution of urban aerosol



Saharan dust outbreaks





The HYSPLIT model

The HYSPLIT model is a complete system for computing simple air parcel trajectories, as well as complex transport, dispersion, chemical transformation, and deposition simulations. HYSPLIT continues to be one of the most extensively used atmospheric transport and dispersion models in the atmospheric sciences community.

The model calculation method is a hybrid between the Lagrangian approach, using a moving frame of reference for the advection and diffusion calculations as the trajectories or air parcels move from their initial location, and the Eulerian methodology, which uses a fixed three-dimensional grid as a frame of reference to compute pollutant air concentrations.

https://www.ready.noaa.gov/HYSPLIT.php

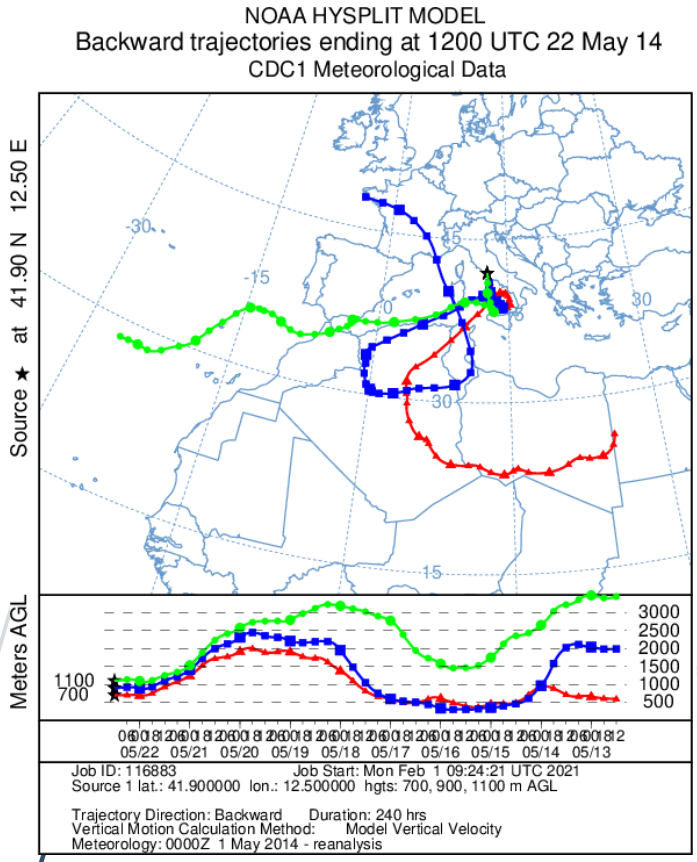
The HYSPLIT model

Meteorological archives available for HYSPLIT:

- GDAS one-degree archive (Dec 2004 - present)
- GFS quarter-degree archive (June 2019 - present)
- NCEP/NCAR Reanalysis (1948 - present)
- HRRR 3 km (June 2019 - present)
- NAM 12 km (May 2007 - present)
- Weather Research & Forecasting Model (WRF ARW, 1980 - 2018)
- …

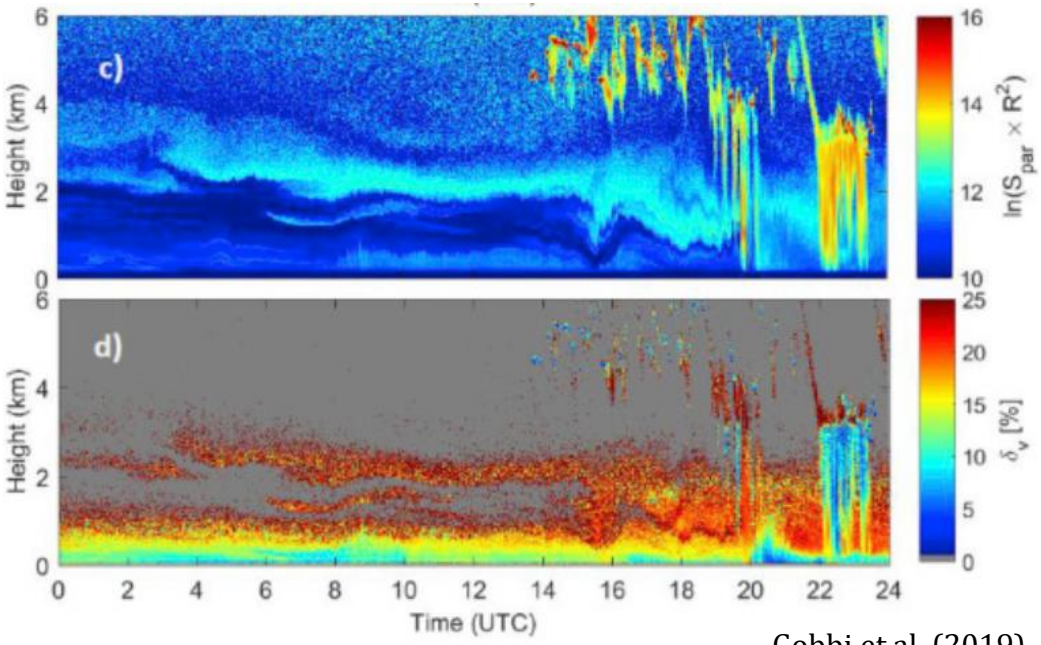
one-degree of latitude ≅ 111 km

The HYSPLIT model - Saharan dust outbreaks



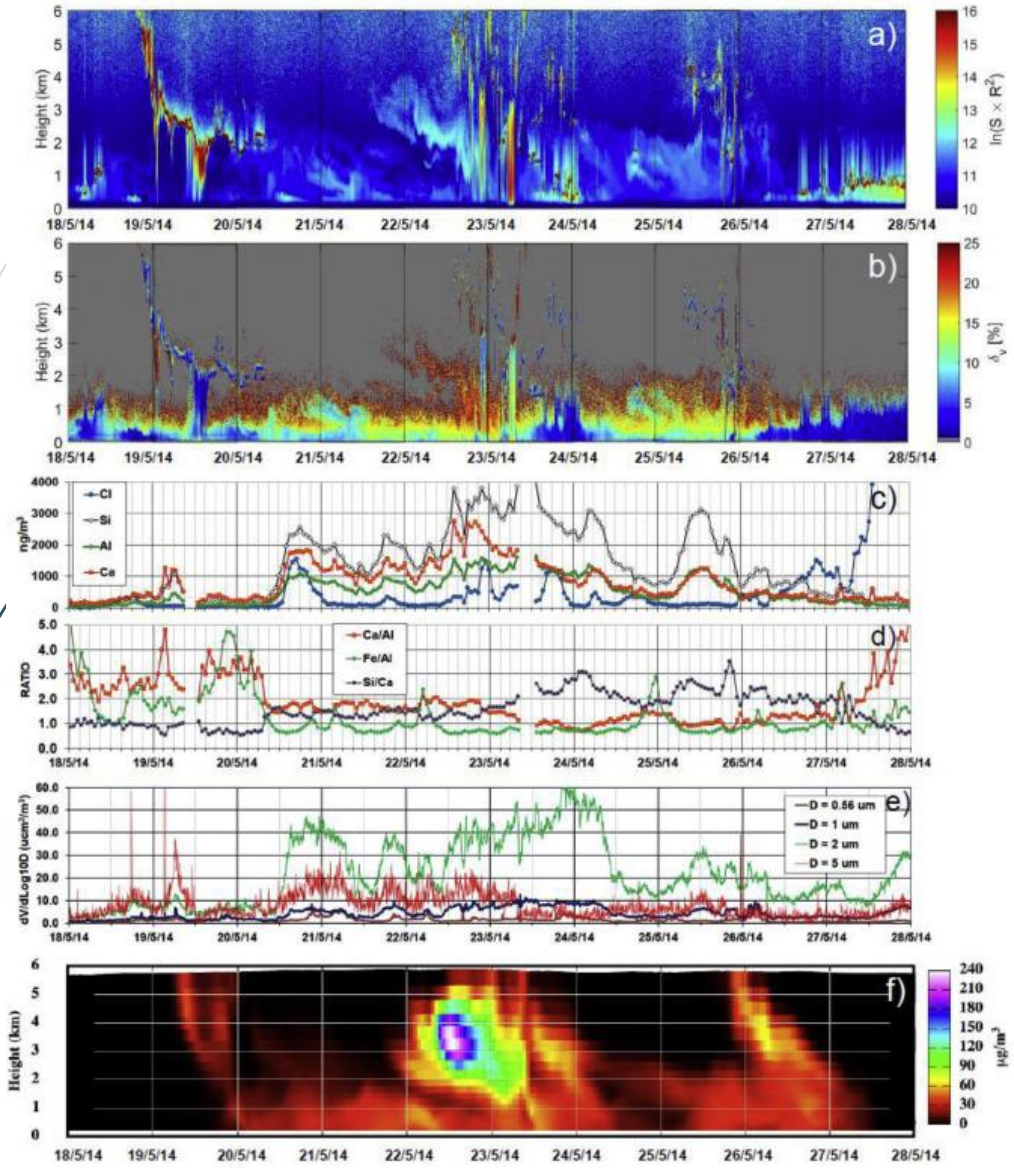
Advection of Saharan dust observed in Rome on May 22, 2014.

Right: 240 hours backtrajectories
Bottom: range-corrected signal ($\ln(S*R^2)$) and depolarization ratio (δ_v%) up to 6 km altitude from ceilometer



Gobbi et al. (2019)

The HYSPLIT model - Saharan dust outbreaks



(a) range-corrected, parallel backscatter signal up to 6 km

(b) volume depolarization ratio

(c) Cl, Si, Al and Ca concentrations in PM_{10}

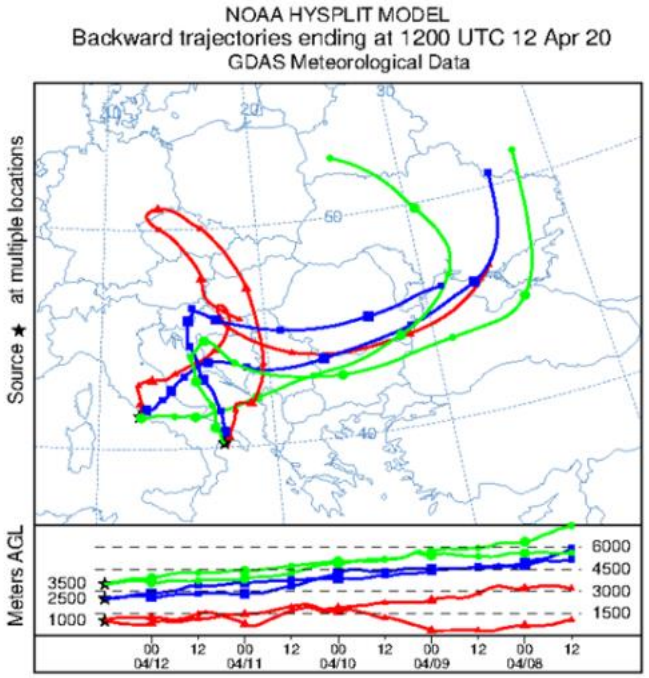
(d) Ca/Al, Fe/Al and Si/Ca ratios of PM_{10} components

(e) aerosol volume distribution (dV/dLogD) at aerodynamic diameters of 0.56, 1, 2 and 5 μm

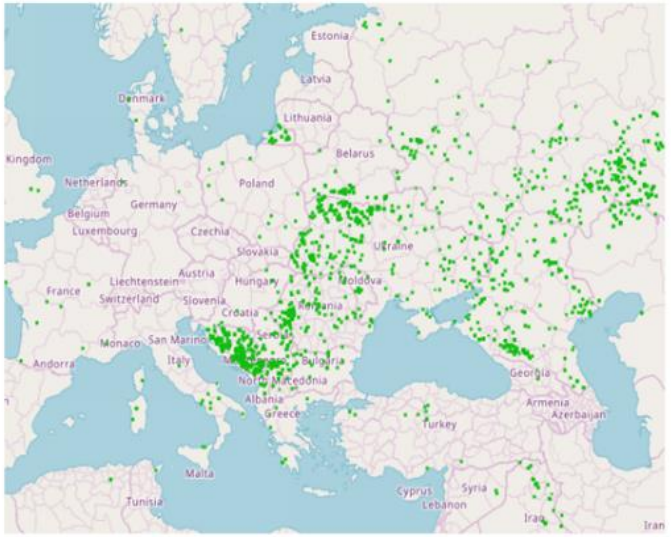
(f) Saharan dust PM_{10} up to 6 km, as simulated by WRF-Chem.

Gobbi et al. (2019)

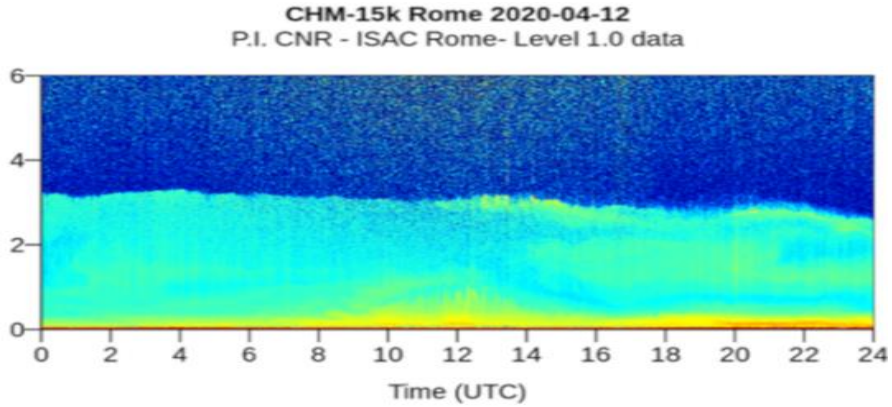
The HYSPLIT model – Fires



96 hours backtrajectories over Rome and Taranto on 12 April, 2020

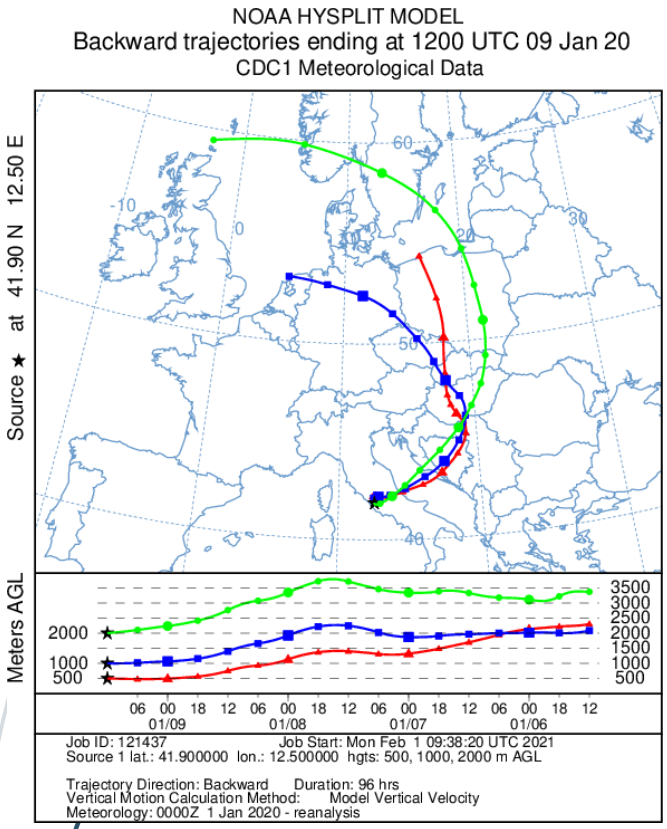


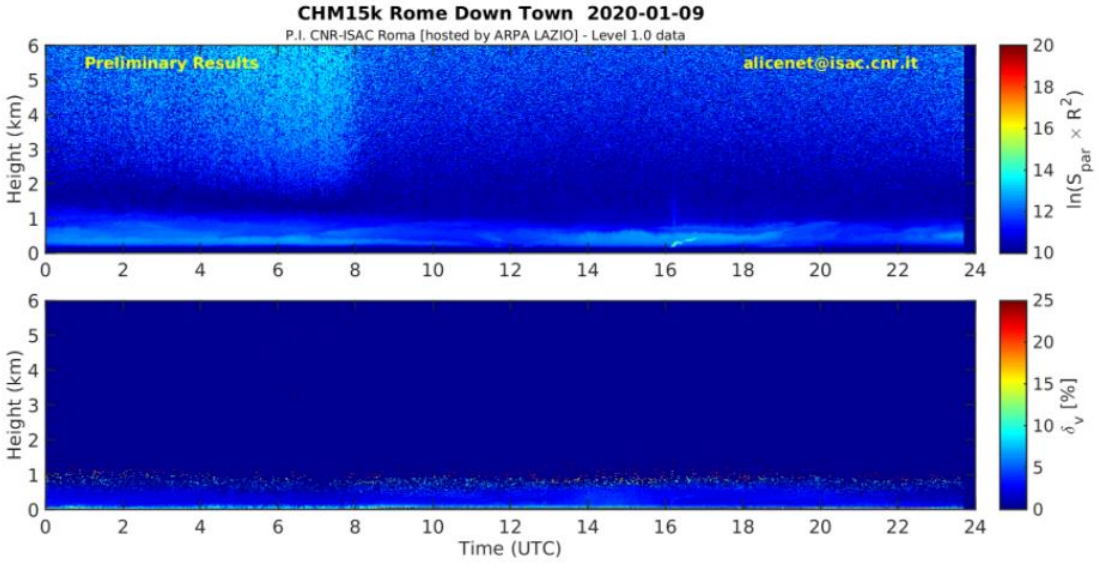
EFFIS active fires on 12 April, 2020



Alice-net backscatter vertical profile over Rome 12 April, 2020

The HYSPLIT model – Continental air



Advection of "clean" continental air observed in Rome on 09 January, 2020.

Right: 96 hours backtrajectories
Bottom: range-corrected signal ($\ln(S*R^2)$) and depolarization ratio (δ_v%) up to 6 km altitude from ceilometer