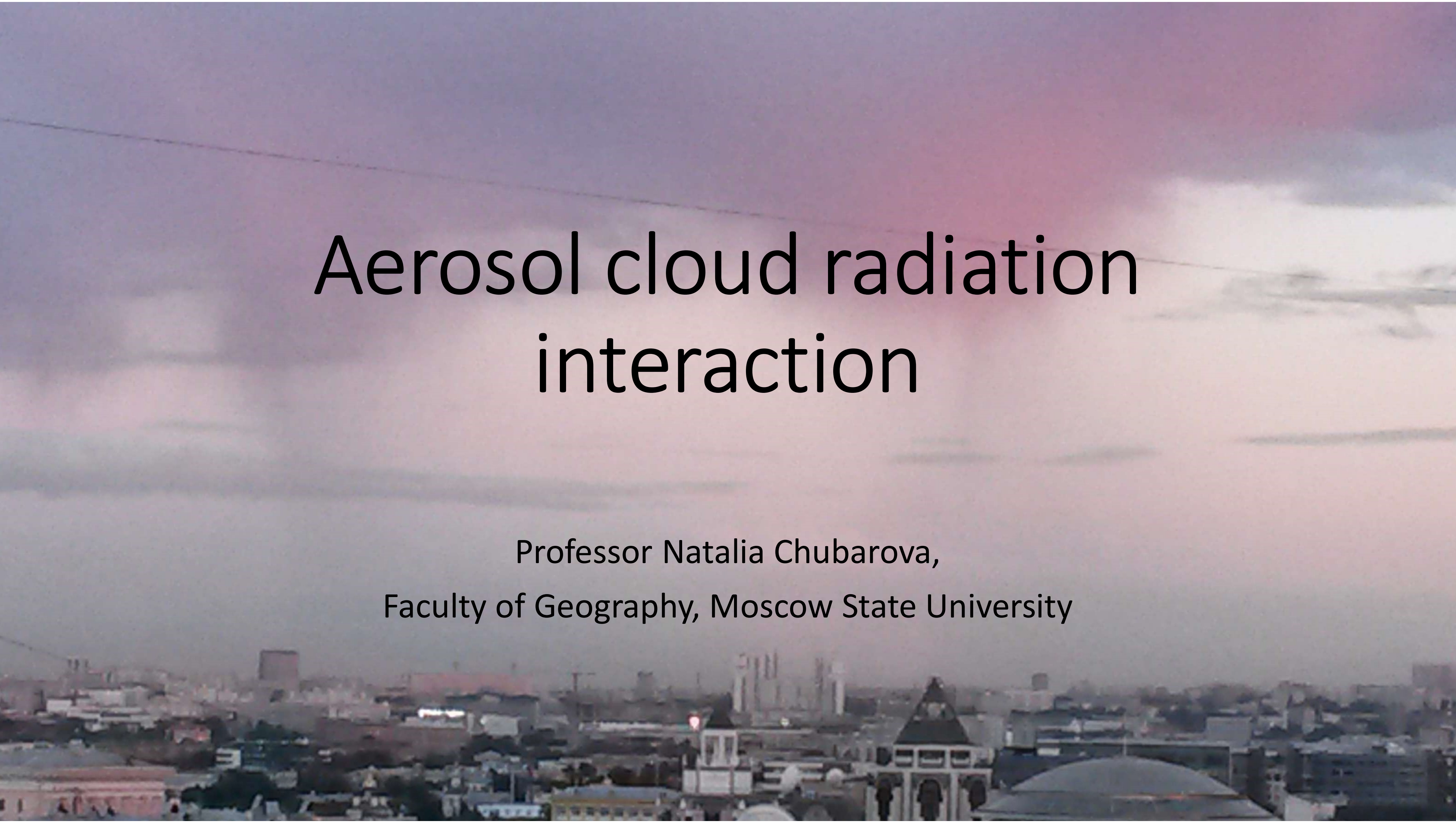


Aerosol cloud radiation interaction

Professor Natalia Chubarova,

Faculty of Geography, Moscow State University

OUTLINE :

1. Solar radiation and main metrics for the description of its interaction with the medium.

2. Aerosol and cloud main characteristics

3. Mechanisms of direct and indirect aerosol effect.

4. Direct cloud and aerosol effects on solar irradiance.

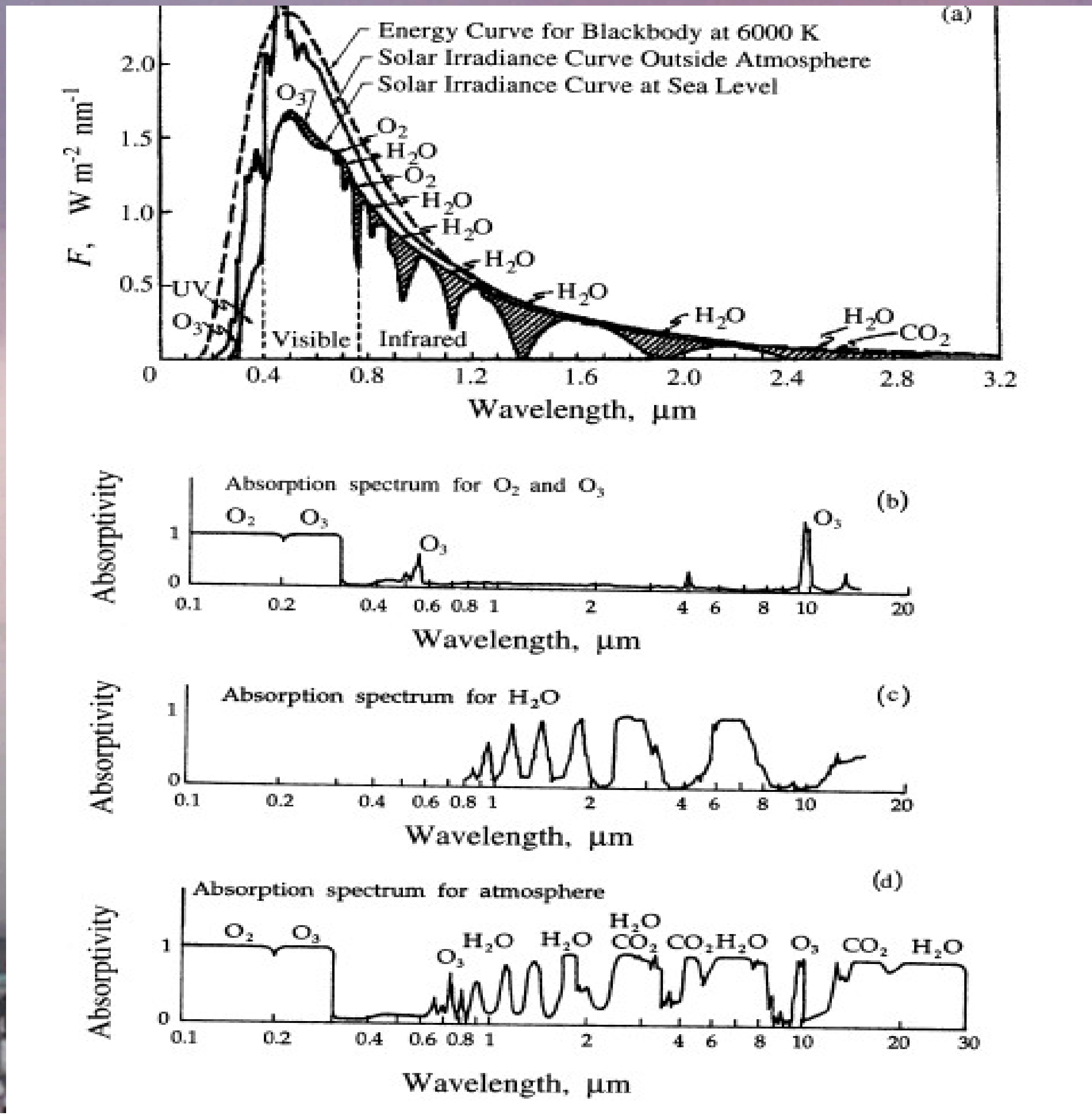
5. Indirect cloud and aerosol effects on solar irradiance.

6. The example of indirect effects during lockdown COVID-19 in Moscow megacity due to aerosol-cloud interaction.

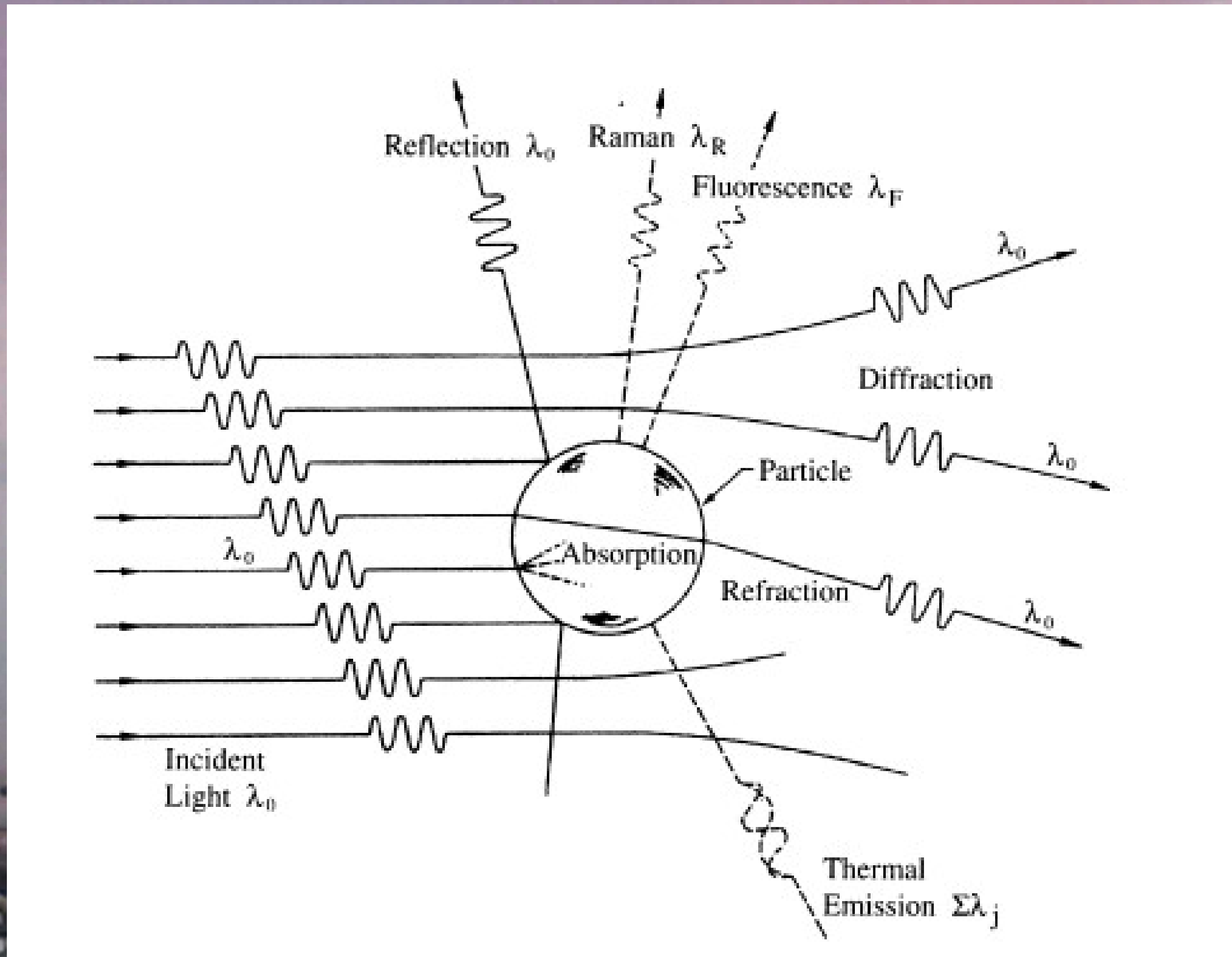
Solar radiation spectrum and absorption bands of various gases

Planck equation:

$$F_B(\lambda) = \frac{2\pi c^2 h \lambda^{-5}}{e^{ch/k\lambda T} - 1}$$

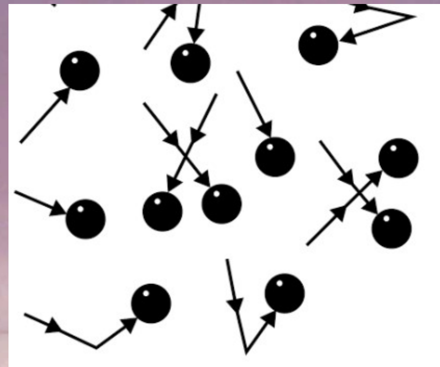


The scheme of radiation interaction with particles



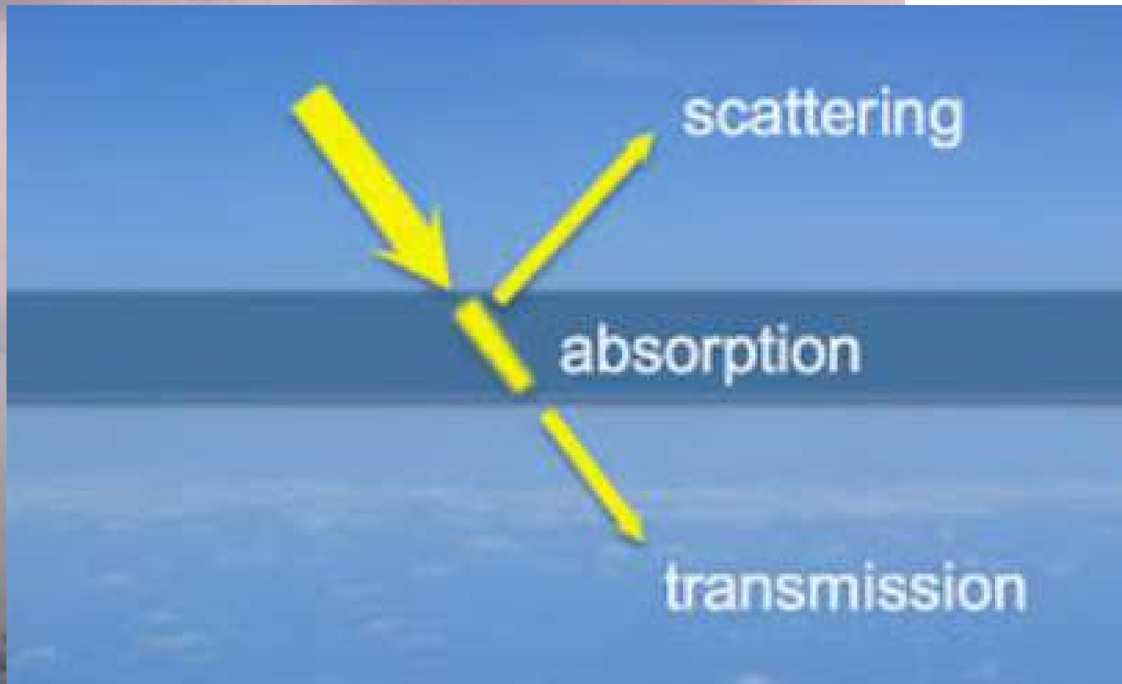
S-P, 2016

Radiative Processes in the Atmosphere



βext = βs + βa

βa – absorption, βs - scattering and
βe extinction coefficients



S-P, 2016

Fundamentals of light scattering in the atmosphere.

Important relations between the size of the radiation wavelength and the radius (structure) of the particle (medium).

It is important to know the particle size and the wavelength of the incident radiation.

$$x = 2\pi r / \lambda$$

Depending on this, several classes of fundamentally different approaches can be used:

1/ $x \ll 1$ ($<10^{-3}$) - neglect of scattering

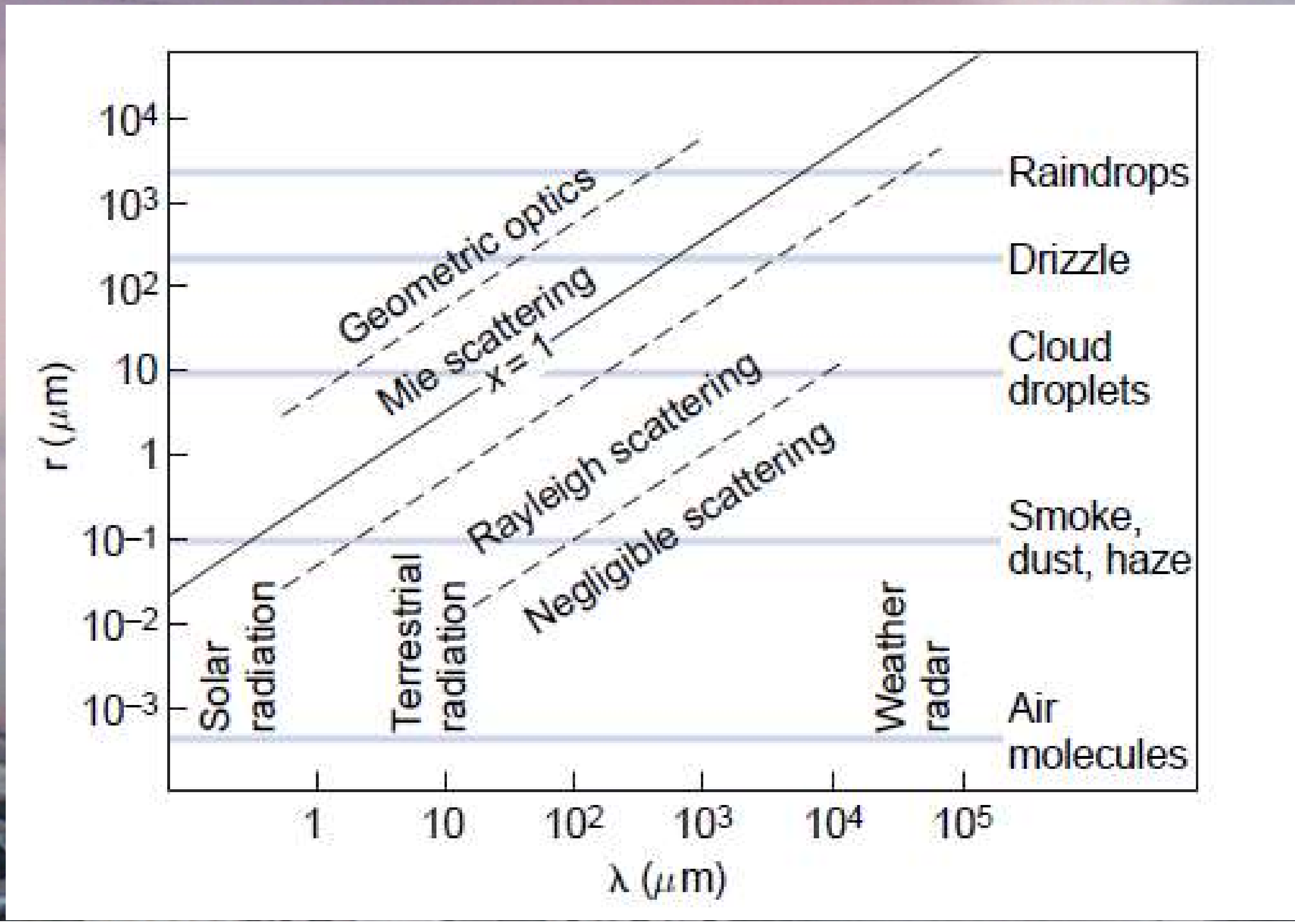
2/ $x = 10^{-3}$-10^{-1} - simplified scattering theory (Rayleigh theory)

3/ $x \sim 10^{-1}$ -50 - representation of scattering in the framework of wave theory - complete scattering theory - theory of Mi

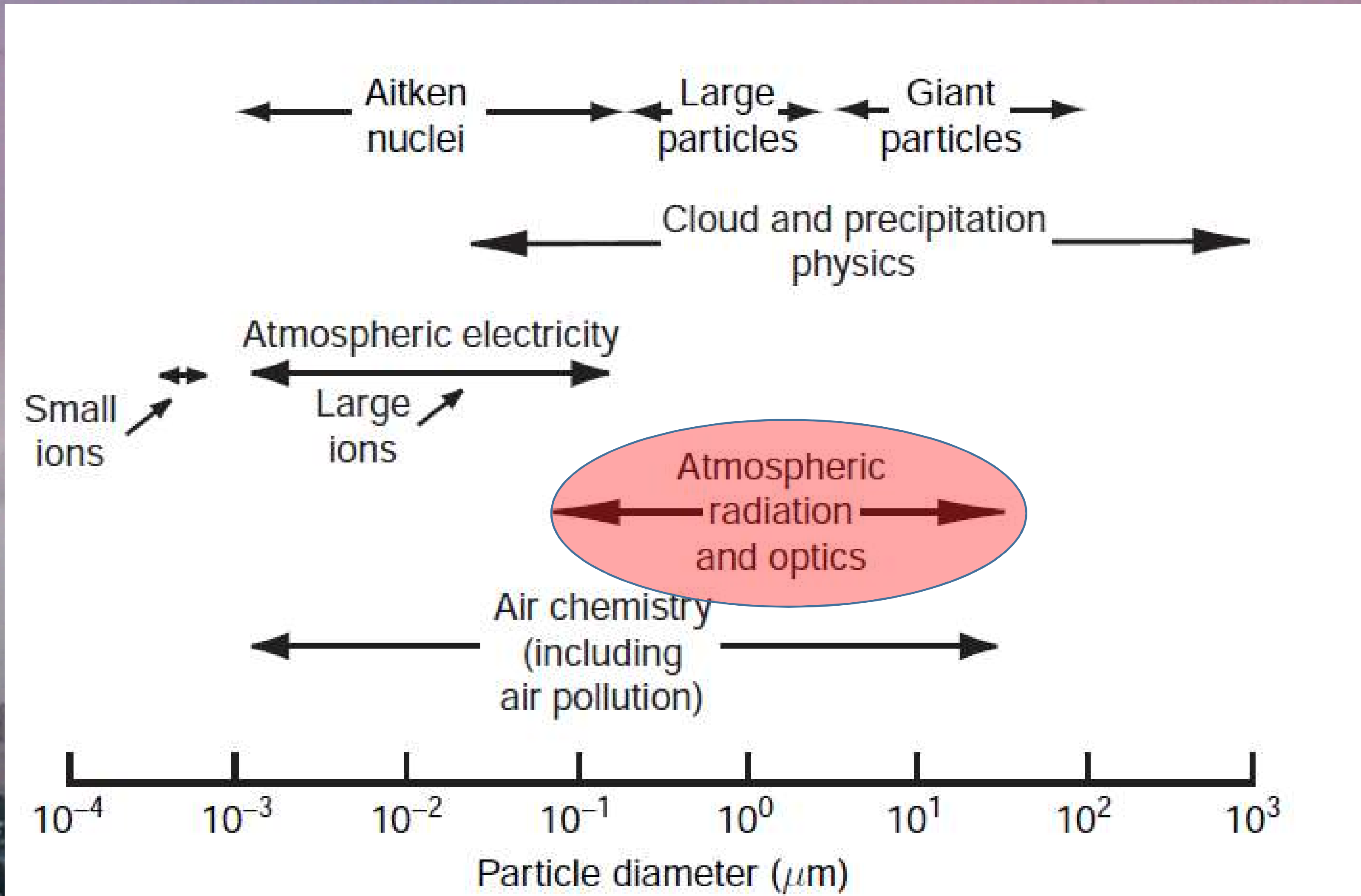
4/ $x > 50$ -geometric optics.

Scattering classes depending on the ratio of wavelength and particle size:

$x=2\pi r/\lambda$



Size range of particles in the atmosphere and their importance.



Wallace Hobbs, 2006

Metrics of main radiative characteristics

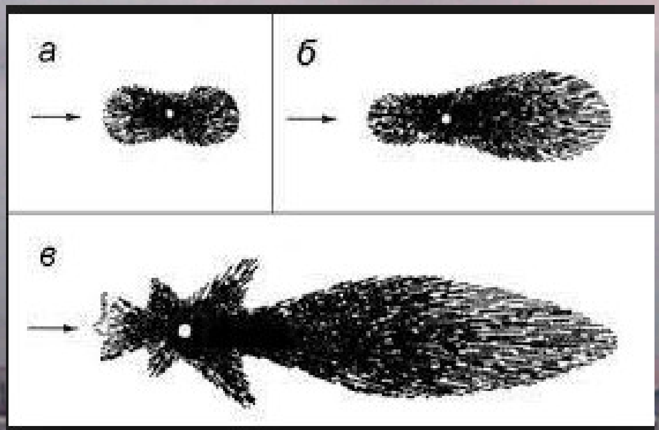
Optical thickness (depth):

$$\tau_\lambda(z_1, z_2) = \int_{z1}^{z2} \beta_{e,\lambda}(z)dz$$

β denotes
βs - scattering and
βe extinction coefficients

Single scattering albedo

$$\varpi_\lambda = \frac{\beta_{s,\lambda}}{\beta_{e,\lambda}}$$

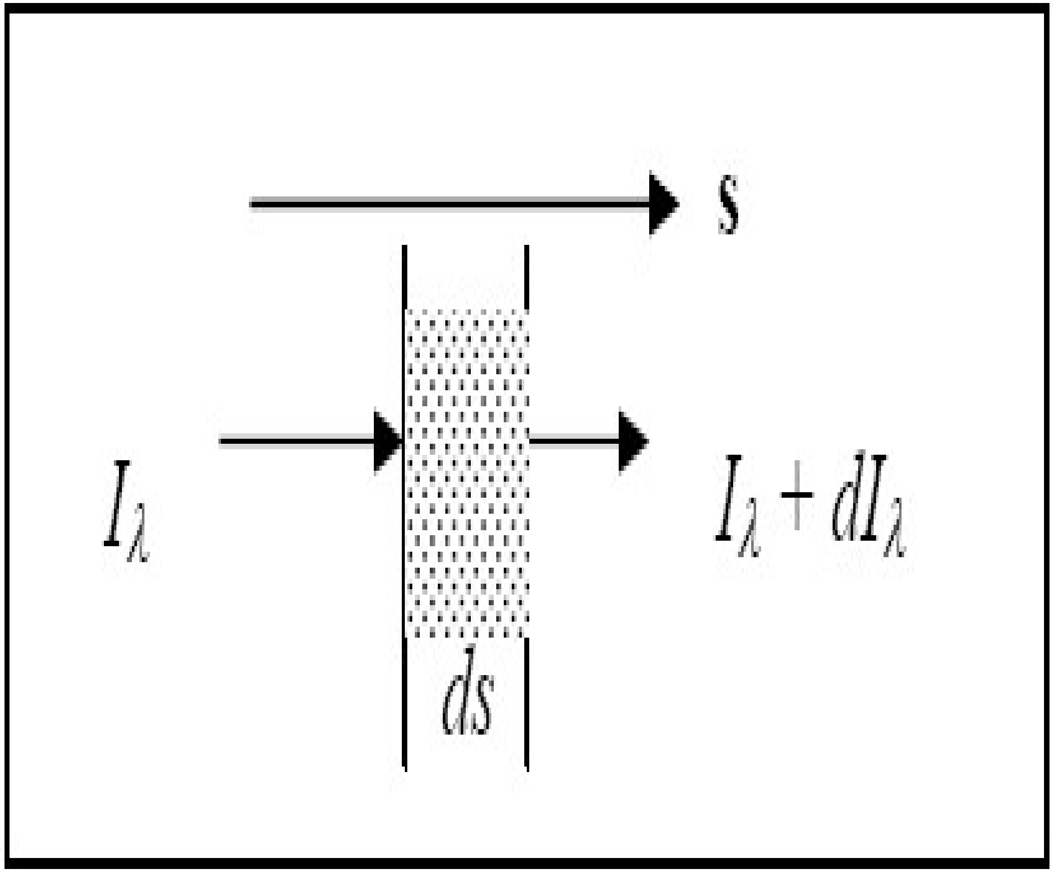


Phase function

$$½ \int P_\lambda(\gamma) \sin\gamma \, d\gamma = 1$$

Asymmetry factor

$$g_\lambda = \frac{1}{2}\int_{-1}^{1} P_\lambda(\cos\gamma)\cos(\gamma)d\cos(\gamma)$$



This is classical Bouguer-Beer - Lambert law :

For attenuation:

$$dI_\lambda = -\beta_{e,\lambda} I_\lambda ds$$

For emission:

$$dI_\lambda = \beta_{e,\lambda} J_\lambda ds$$

In case of both emission and absorption the equation is the following:

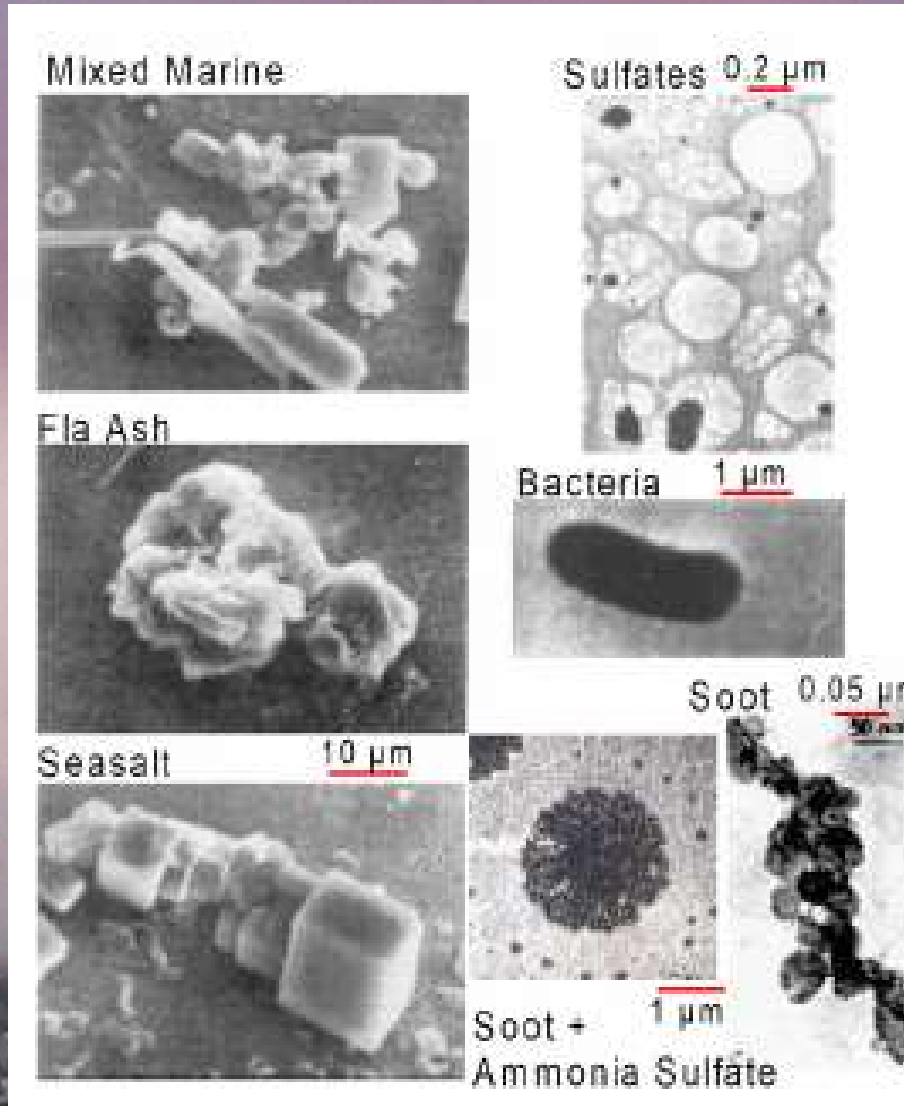
$$dI_\lambda = -\beta_{e,\lambda} I_\lambda ds + \beta_{e,\lambda} J_\lambda ds$$

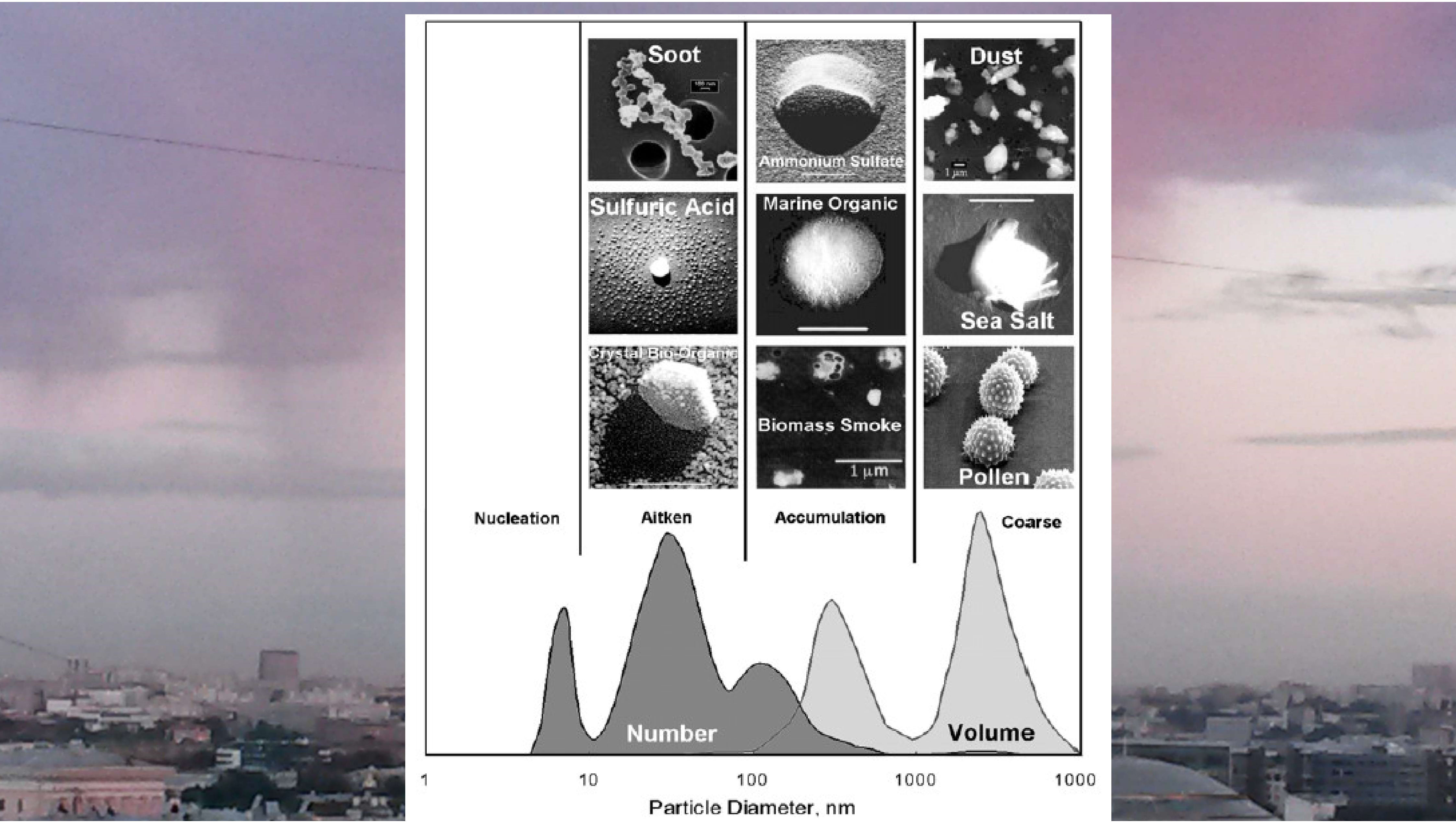
This is the differential form of radiative transfer equation.

2. Aerosol and cloud : main characteristics

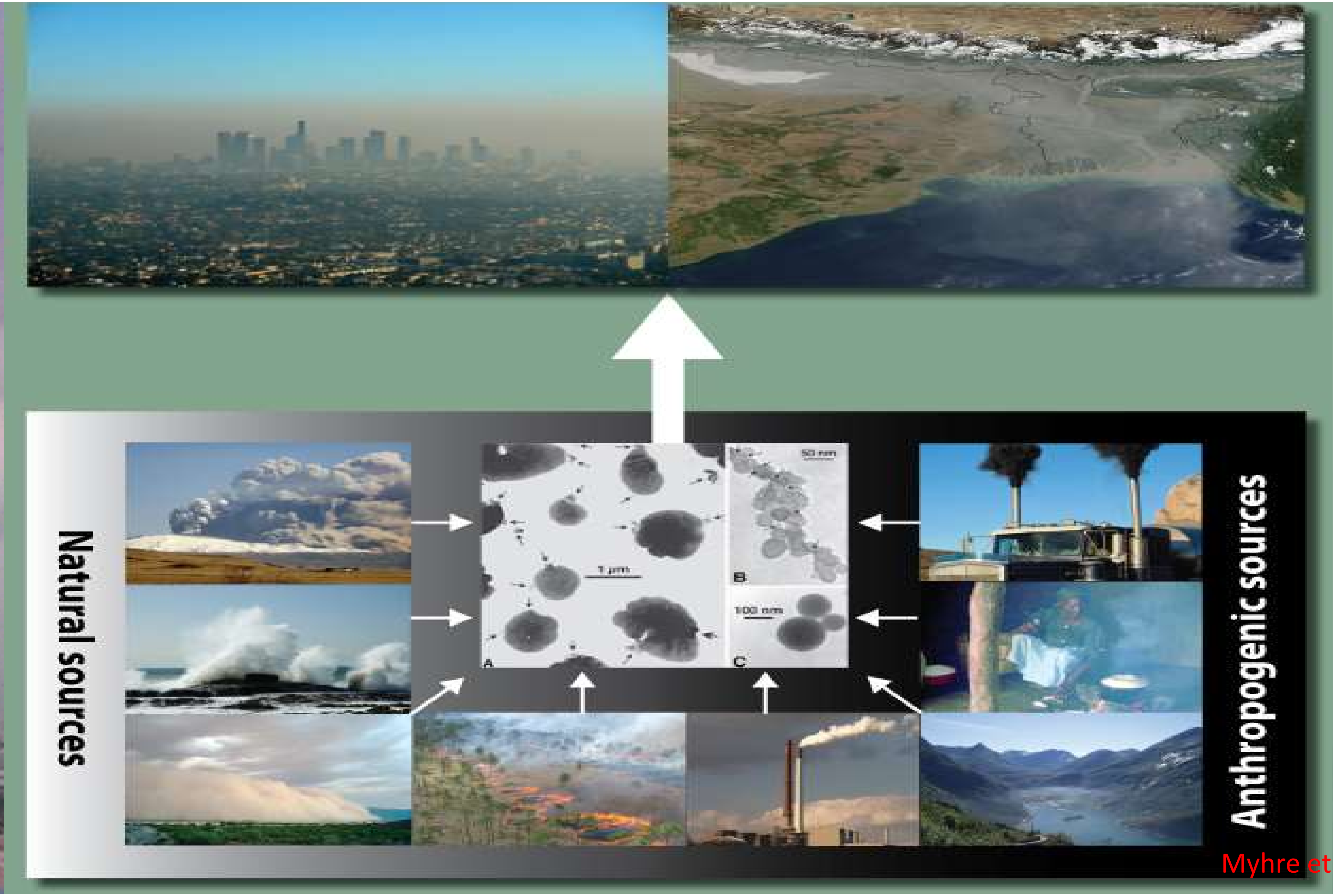
<u>Atmospheric aerosols</u> are suspensions of small particles in the atmosphere with diameter of 0.001 to 100 micron.

We need to know their
microphysical (size distribution)
optical (refractive index) properties
for obtaining their <u>radiative properties -</u>
[spectral aerosol optical thickness (depth), phase function and single scattering albedo.]





Soot
Ammonium Sulfate
Dust
1 µm
Sulfuric Acid
Marine Organic
Sea Salt
Crystal Bio-Organic
Biomass Smoke
1 µm
Pollen
Nucleation
Aitken
Accumulation
Coarse
Number
Volume
1
10
100
1000
1000
Particle Diameter, nm



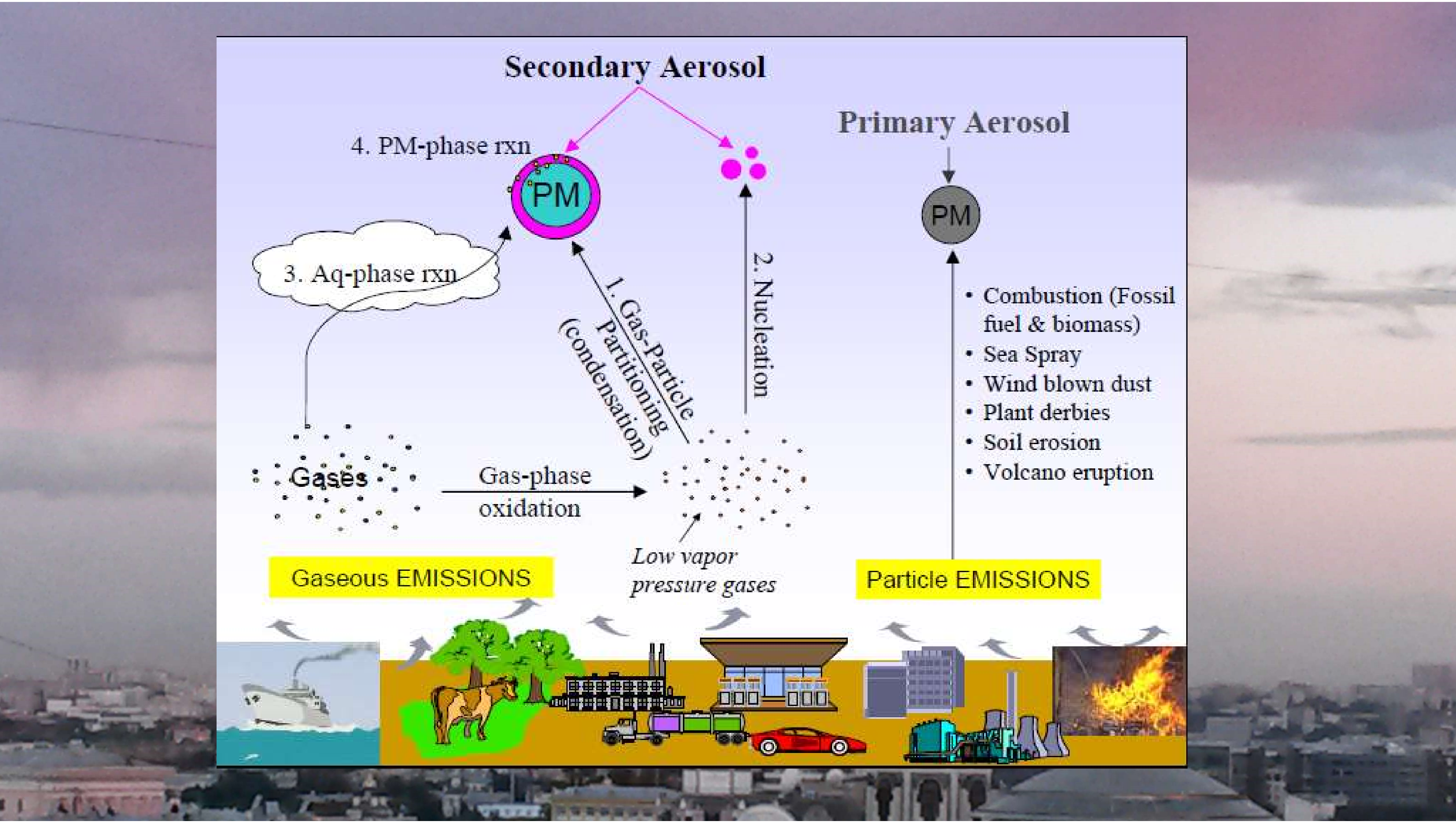
Myhre et al., 2013

Moscow

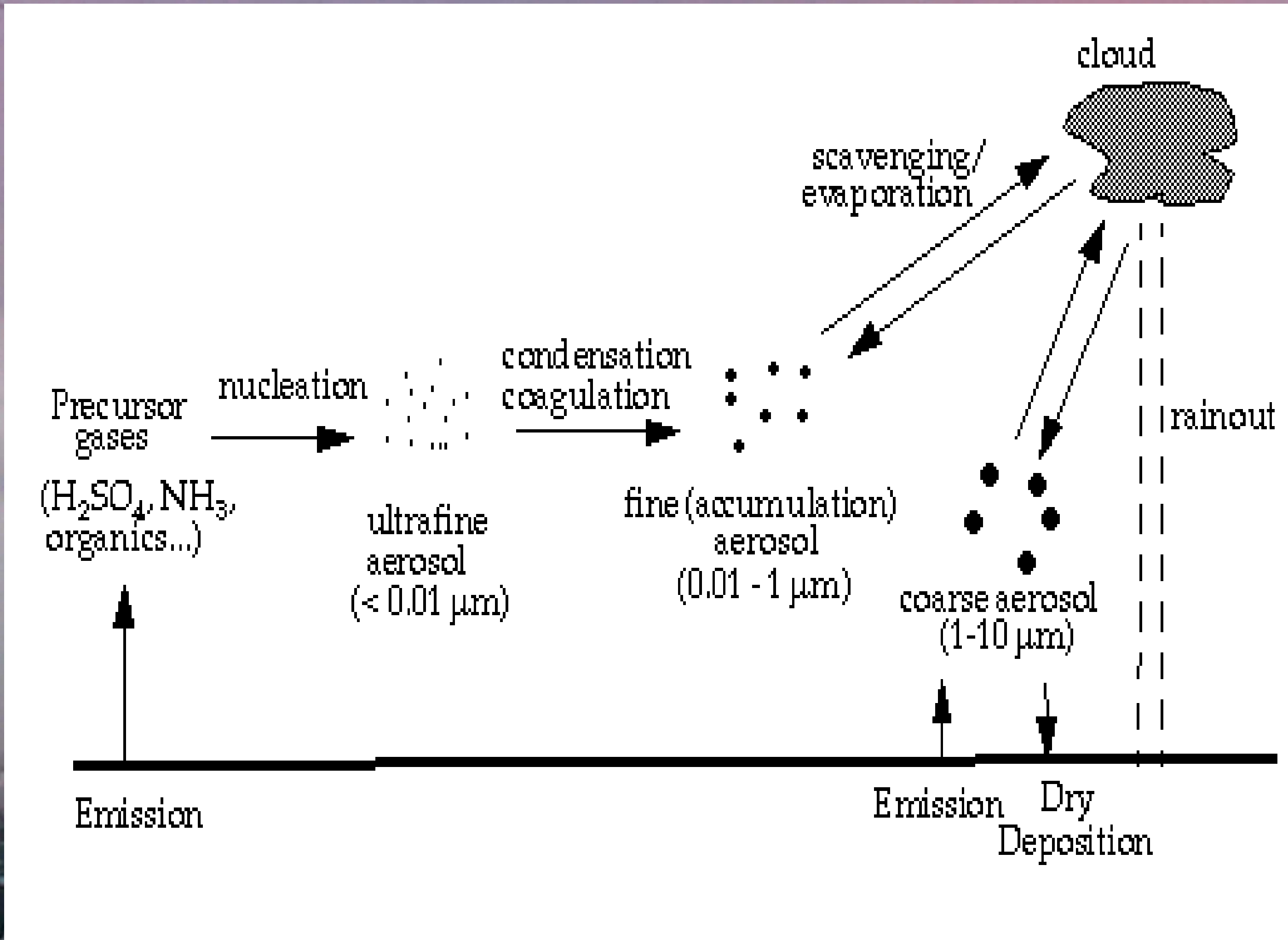
Smoke aerosol, 2002

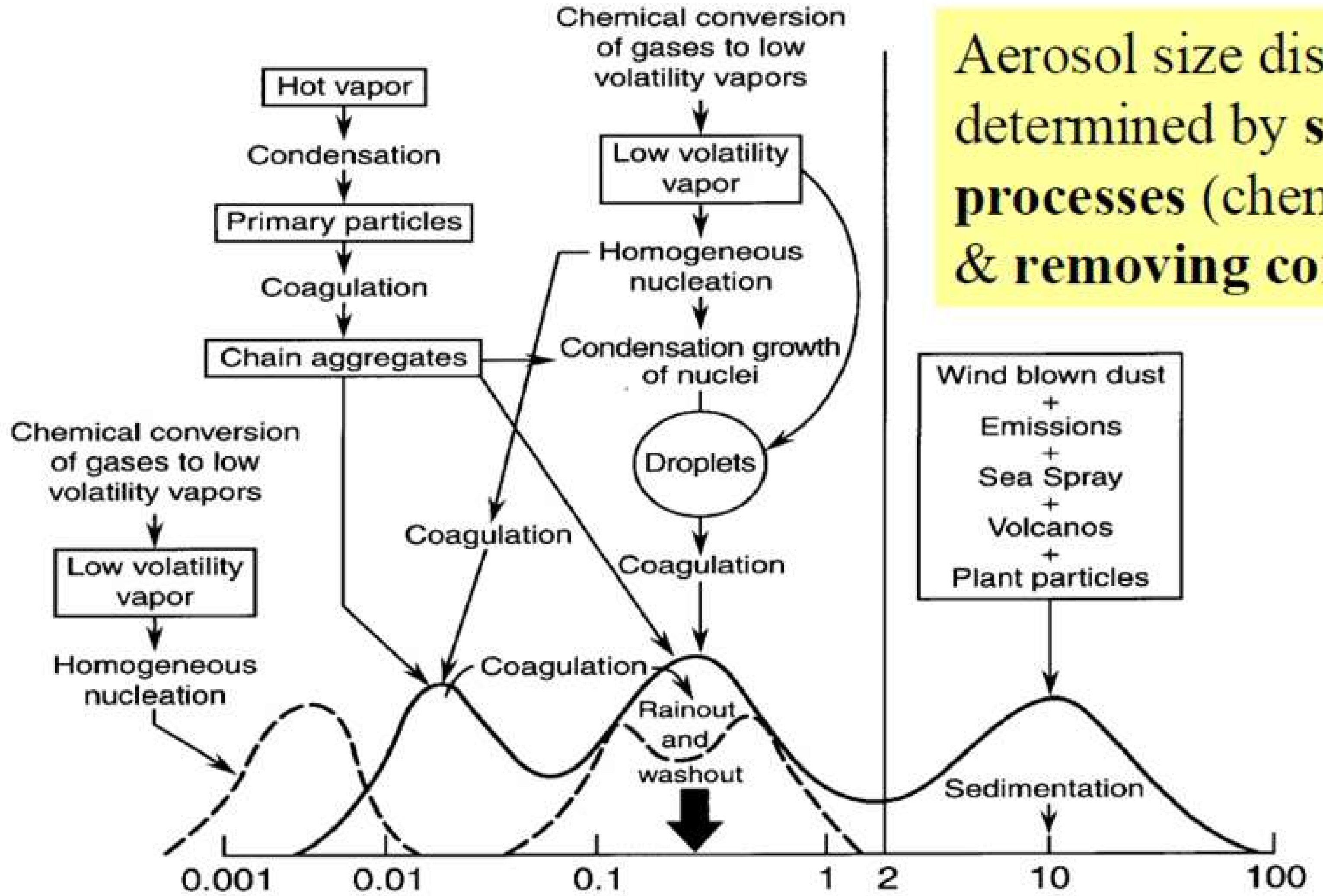
Typical urban aerosol

Chubarova, 2009


Secondary Aerosol
Primary Aerosol
4. PM-phase rxn
PM
PM
3. Aq-phase rxn
1. Gas-Particle Partitioning (condensation)
2. Nucleation
• Combustion (Fossil fuel & biomass)
• Sea Spray
• Wind blown dust
• Plant derbies
• Soil erosion
• Volcano eruption
Gases
Gas-phase oxidation
Low vapor pressure gases
Gaseous EMISSIONS
Particle EMISSIONS

Production, growth, and removal of atmospheric aerosols





Aerosol size distributions are determined by **source**, **processes** (chemical, physical), & **removing conditions**.

Finlayson-Pitts &Pitts

- Estimates (in Tg per year) for the year 2000 of
- (a) direct particle emissions into the atmosphere and (b) *in situ*
- production

(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

IPCC 2007

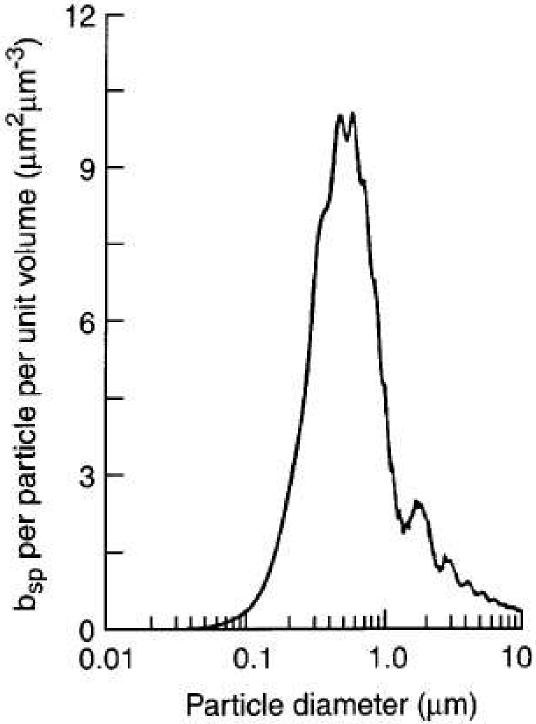


FIGURE 9.22 Scattering coefficient per particle divided by particle volume plotted as a function of diameter. The particles are assumed to be spheres of refractive index 1.50 and the light has $\lambda = 550$ nm (adapted from Waggoner and Charlson, 1976).

Finlayson-Pitts & Pitts

The distribution of aerosol optical thickness over the world

$$\tau_{\lambda}(z_1, z_2) = \int_{z1}^{z2} \beta_{e,\lambda}(z)dz$$

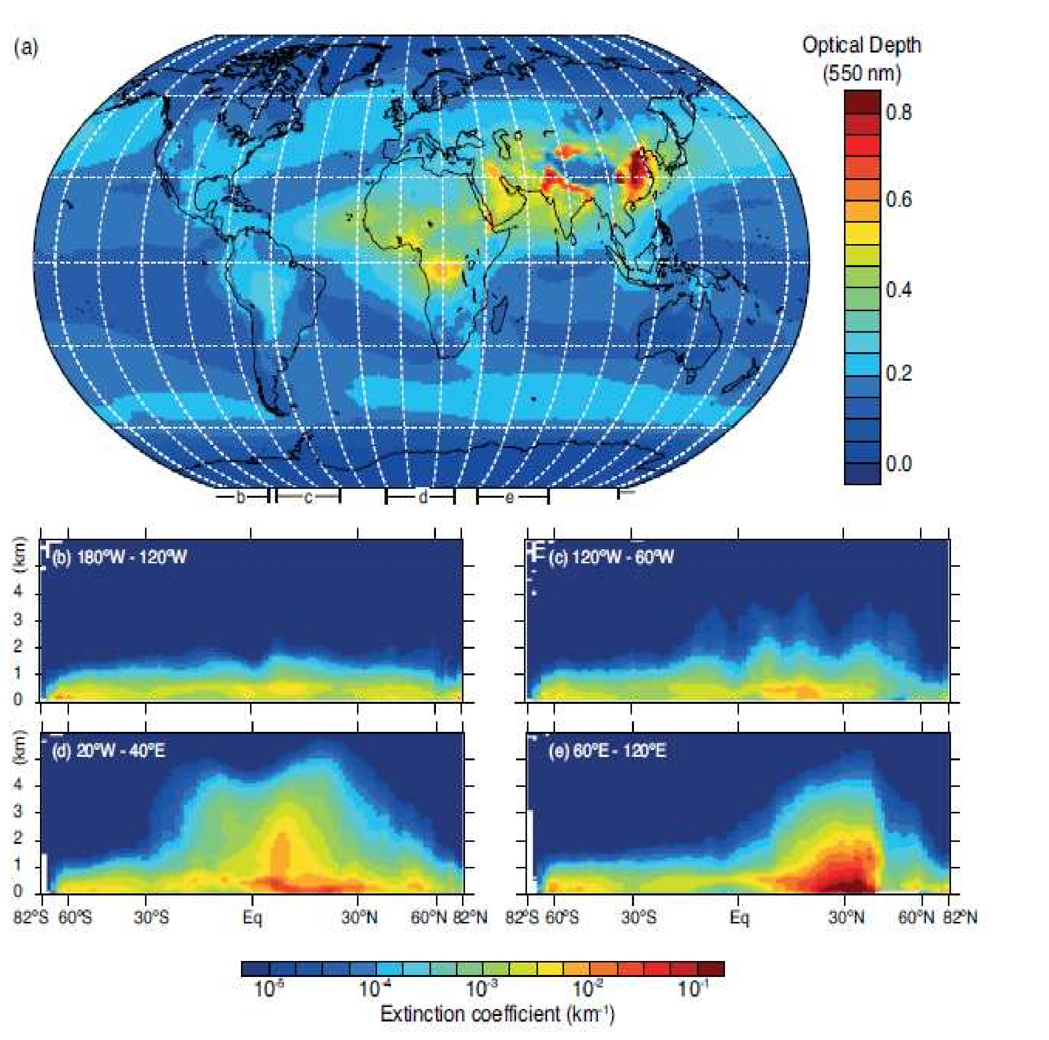
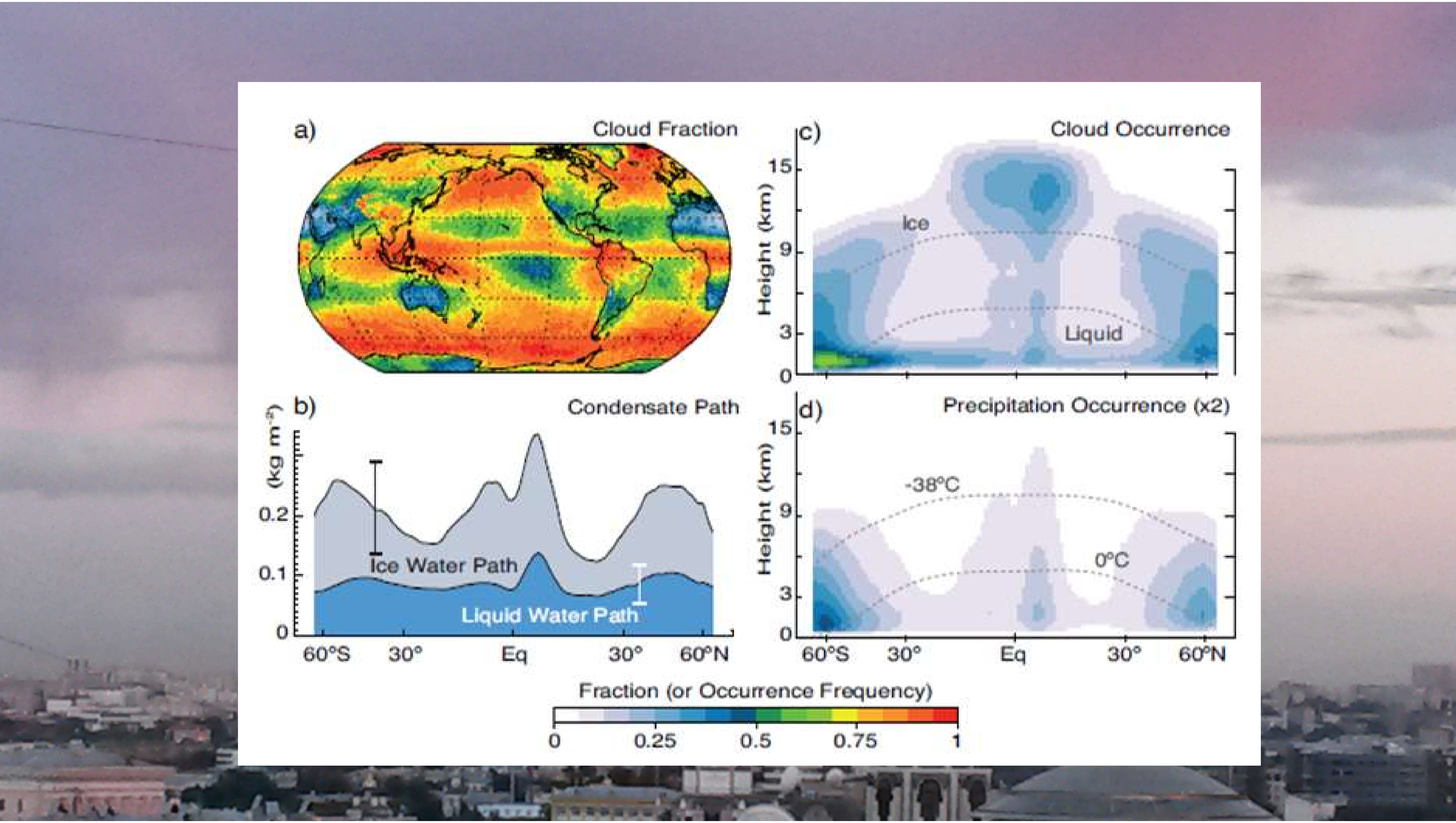


Figure 7.14 | (a) Spatial distribution of the 550 nm aerosol optical depth (AOD, unitless) from the European Centre for Medium Range Weather Forecasts (ECMWF) Integrated Forecast System model with assimilation of Moderate Resolution Imaging Spectrometer (MODIS) aerosol optical depth (Benedetti et al., 2009; Morcrette et al., 2009) averaged over the period 2003–2010; (b–e) latitudinal vertical cross sections of the 532 nm aerosol extinction coefficient (km^{-1}) for four longitudinal bands (180°W to 120°W, 120°W to 60°W, 20°W to 40°E, and 60°E to 120°E, respectively) from the Cloud–Aerosol Lidar with Orthogonal Polarization (CALIOP) instrument for the year 2010 (nighttime all-sky data, version 3; Winker et al., 2013).

<u>Cloud particles</u> is an aqueous aerosol suspended in the atmosphere with particle sizes from 0.5 (smaller are condensation nuclei) to 1000 microns.

Unlike a non-aqueous aerosol, it is characterized by a more complex spatial structure.



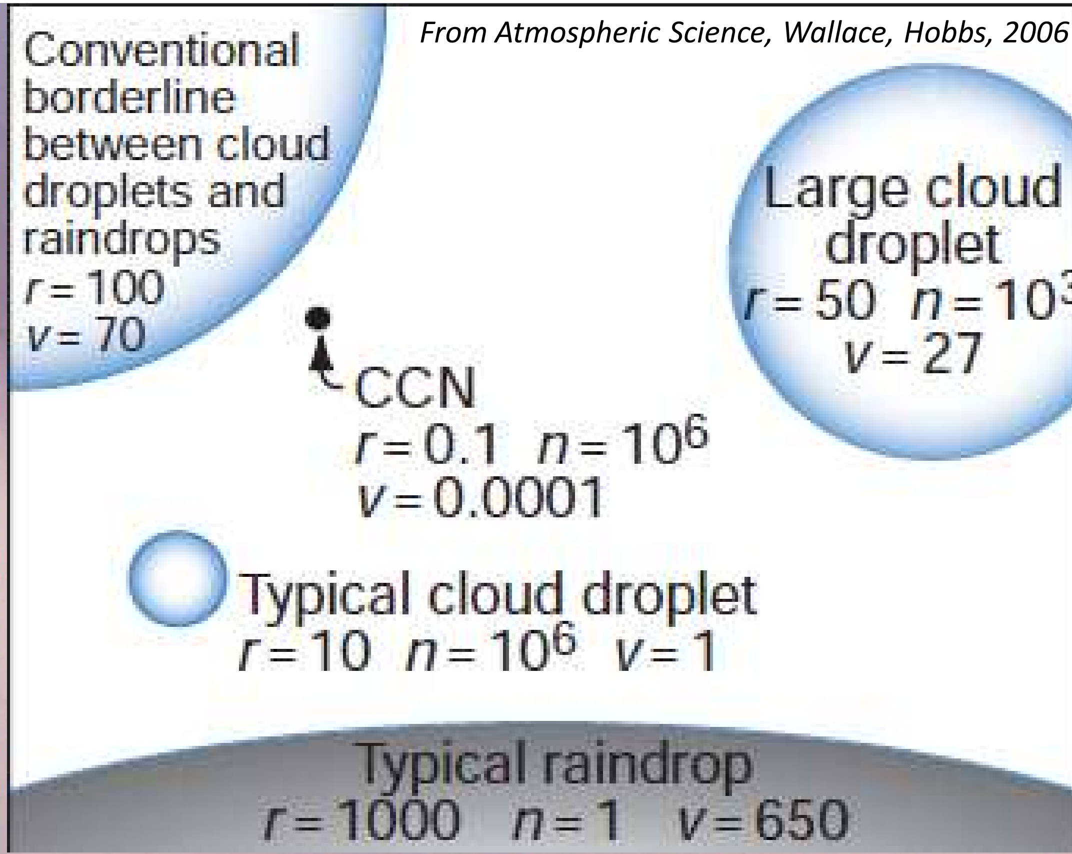
a)
Cloud Fraction
b)
Condensate Path
(kg m⁻²)
0.2
0.1
0
Ice Water Path
Liquid Water Path
60°S
30°
Eq
30°
60°N
c)
Cloud Occurrence
Height (km)
15
9
3
0
Ice
Liquid
d)
Precipitation Occurrence (x2)
Height (km)
15
9
3
0
-38°C
0°C
60°S
30°
Eq
30°
60°N
Fraction (or Occurrence Frequency)
0
0.25
0.5
0.75
1

PROCESSES PROMOTING THE GROWTH OF CLOUD DROPLETS:

Nucleation – formation of particles via self-assembly or self-organization

Coagulation is the unification of small dispersed systems into larger ones - (Brownian, gravitational, turbulent, electrostatic);

Condensation (transition to liquid or solid state from gaseous)



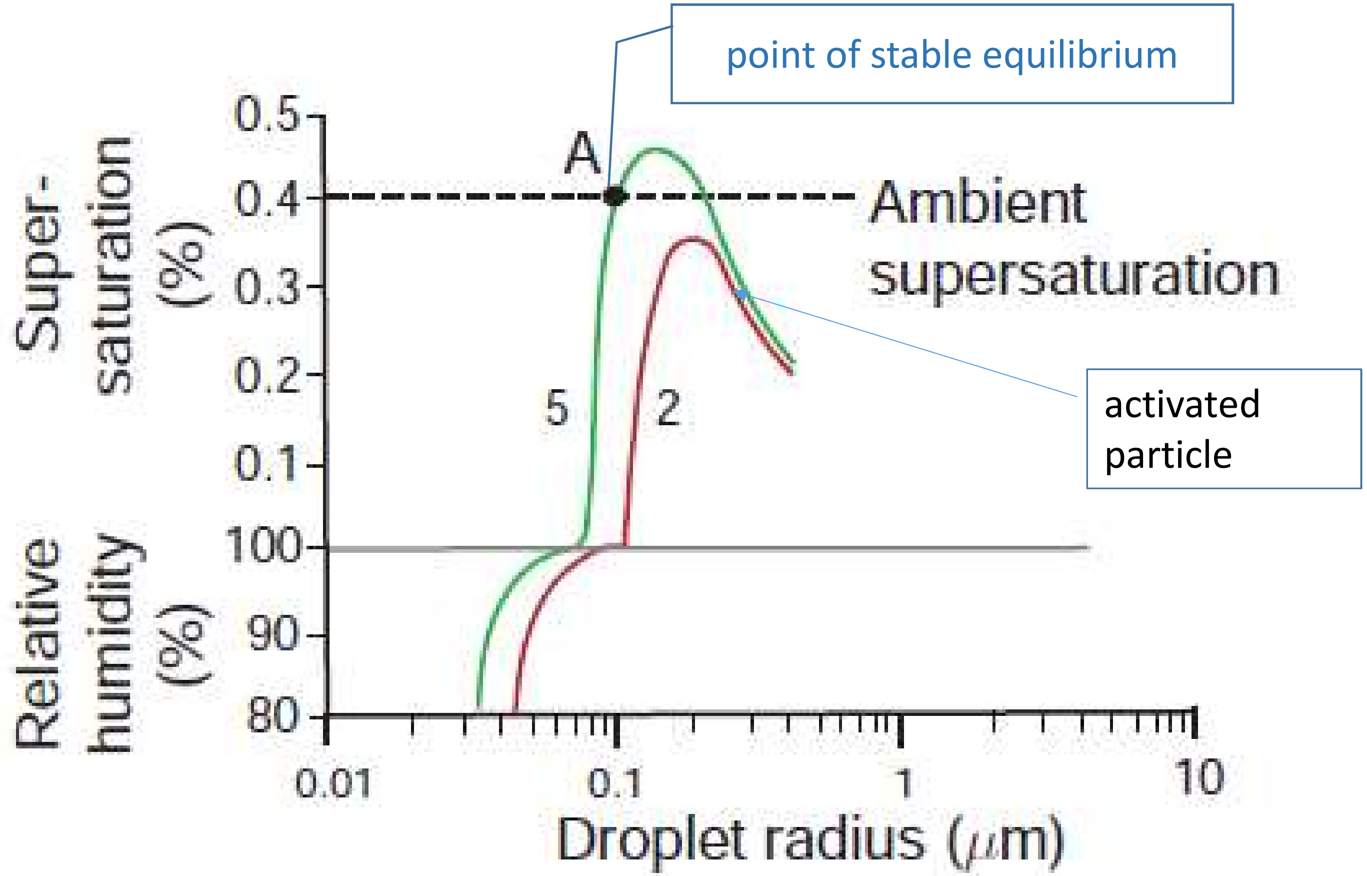
From Atmospheric Science, Wallace, Hobbs, 2006

Cloud condensation nuclei (CCNs) are **small particles typically 0.2 μm, on which water vapor condenses**. Water requires a non-gaseous surface to make the transition from a vapor to a liquid; this process is called condensation

Most CCN consist of a mixture of soluble and insoluble components. The bulk hygroscopicity parameter k has been introduced as a concise measure of how effectively an aerosol particle acts as a CCN.

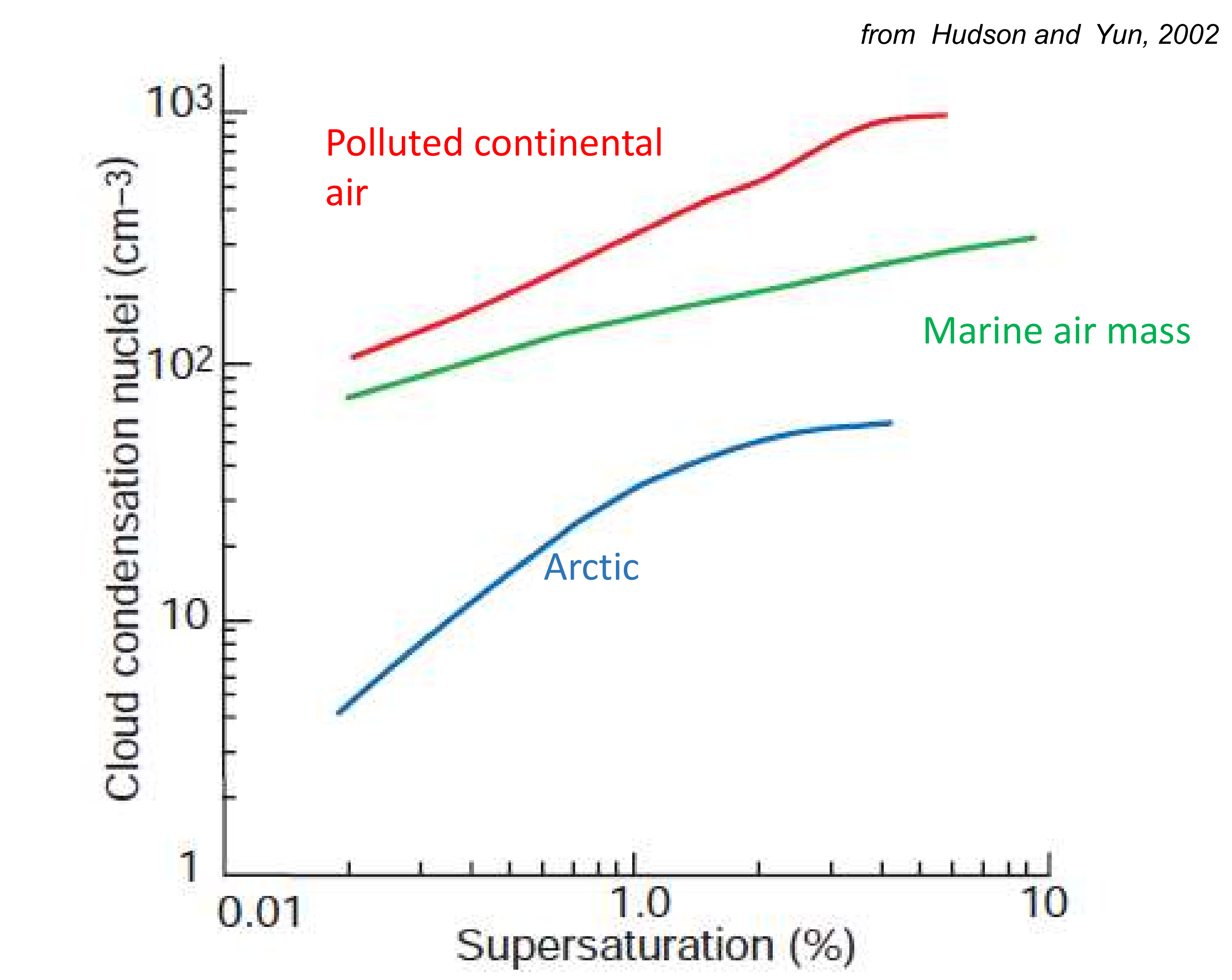
With the increase of the particle size more active processes of its wetting by water are taken place, which in turn lead to the decrease in supersaturation at which the particle can serve as a CCN.

Variations of the relative humidity and supersaturation adjacent to solution droplets containing the following fixed masses of salt: (2) 10^{19} kg of NaCl, (5) 10^{19} kg of $(NH_4)_2SO_4$,



From Atmospheric Science, Wallace, Hobbs, 2006

Concentration of cloud condensation nuclei as a function of supersaturation in different air mass.

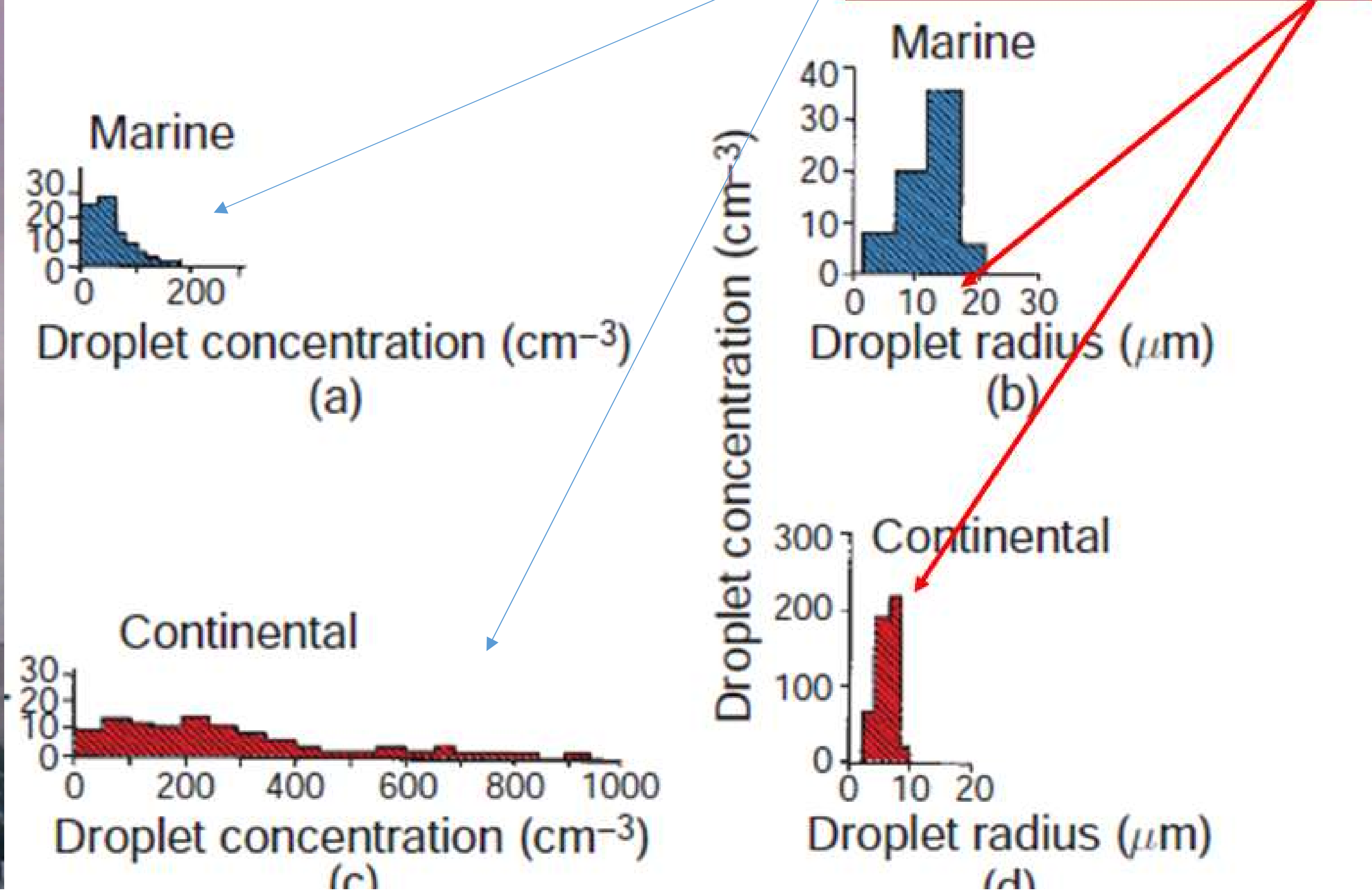


Sulphate particles are the most important source of CCN.
Natural source:
Dimethyl sulfide (DMS) from organic Sulphur and
Methane sulfonic acid (MSA).

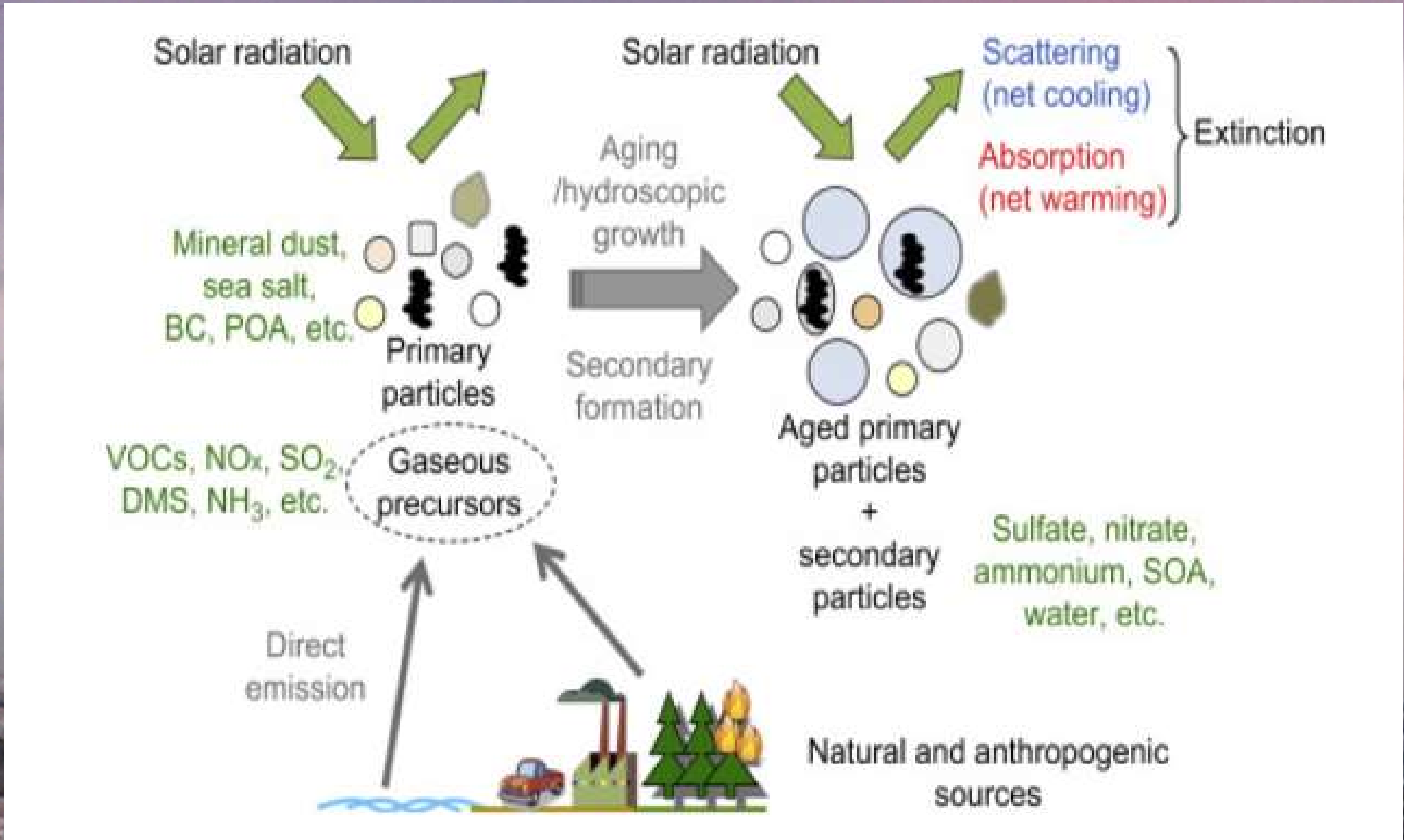
Evaporating clouds also release sulfate particles.

Marine and continental clouds: frequency histogram of droplet concentration and droplet radius

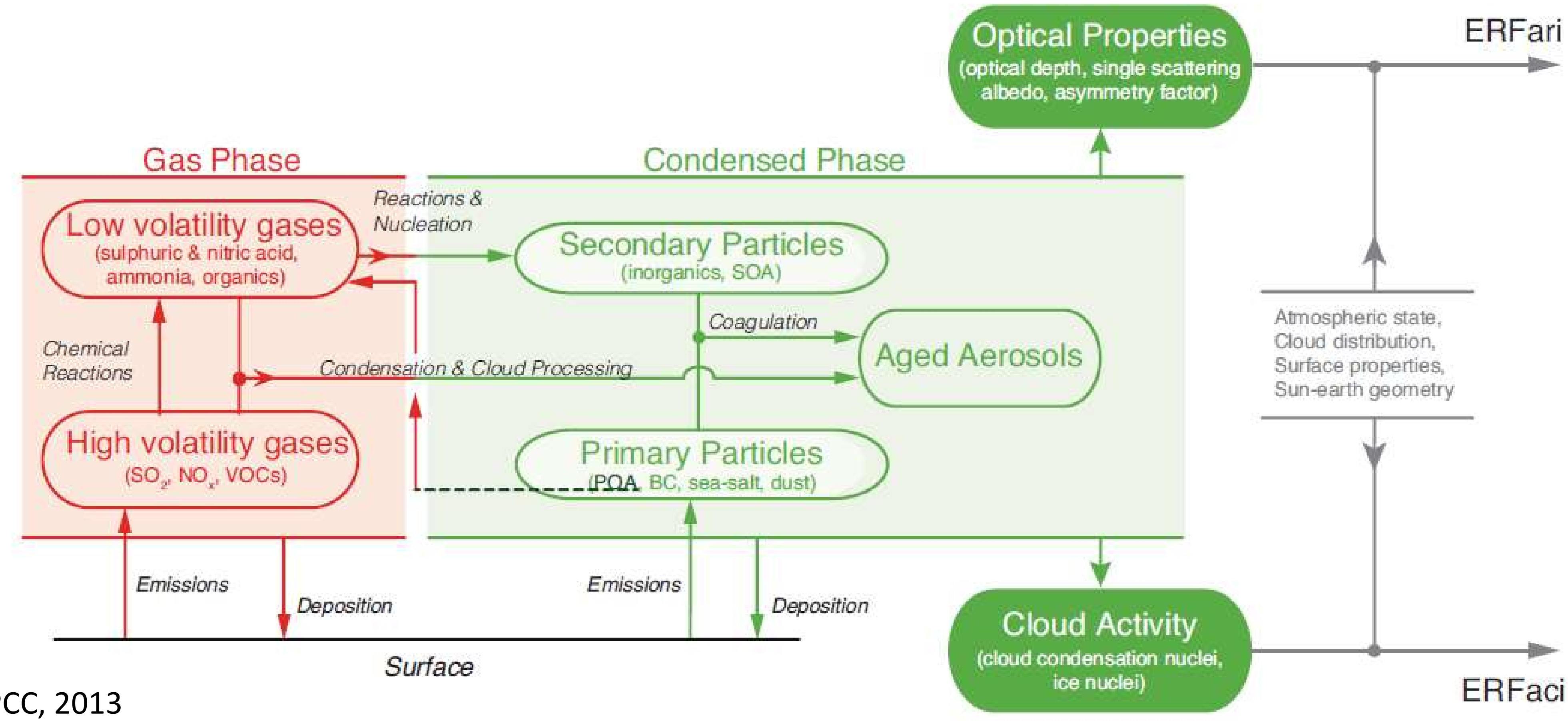
Just remember this important fact!!



3. Mechanisms of direct and indirect aerosol effect.



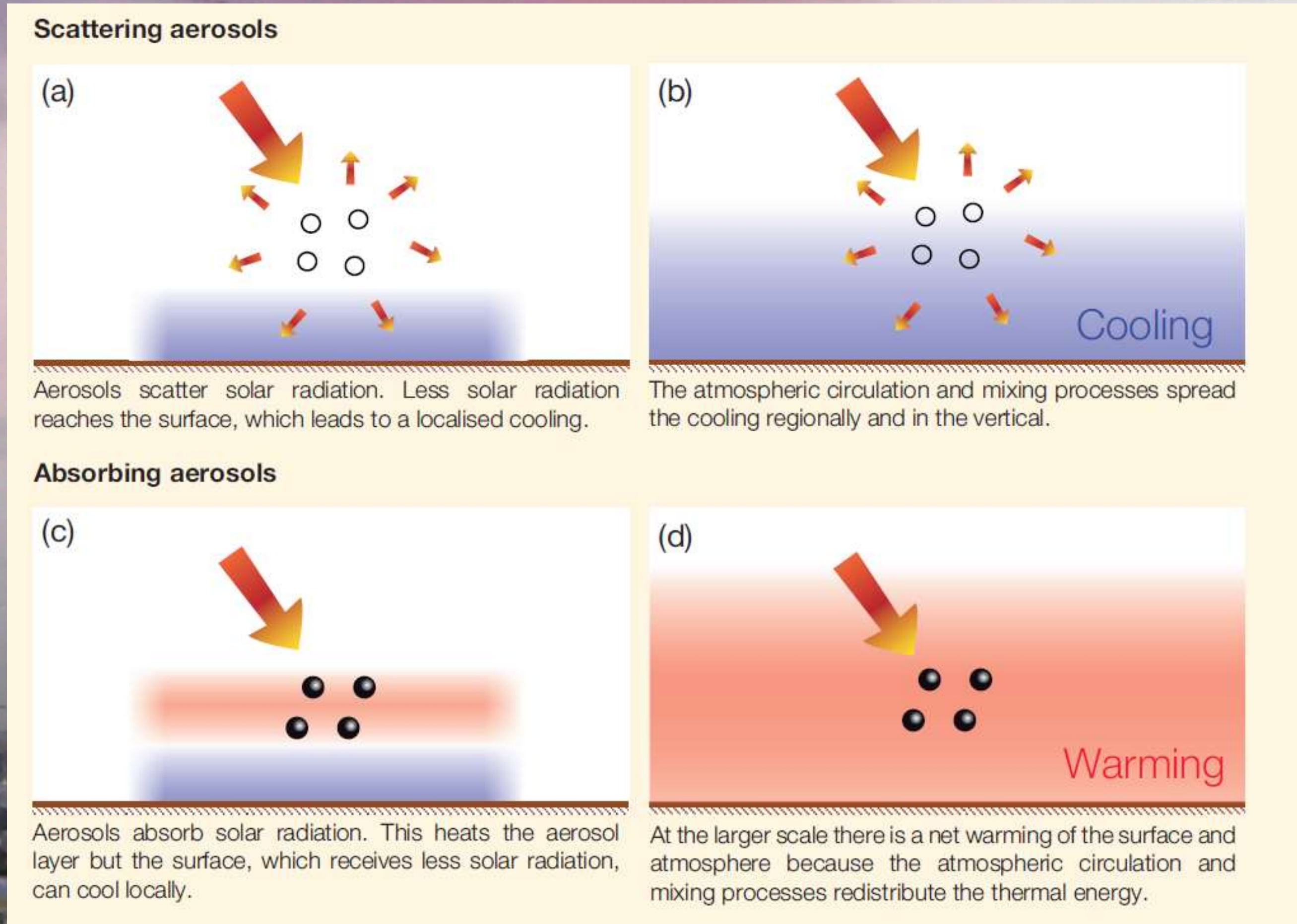
Atmospheric aerosol and environmental variables and processes influencing aerosol–radiation and aerosol–cloud interactions. (ERFari and ERFaci), it is increasingly recognized that aerosols and clouds form a coupled system with two-way interactions.



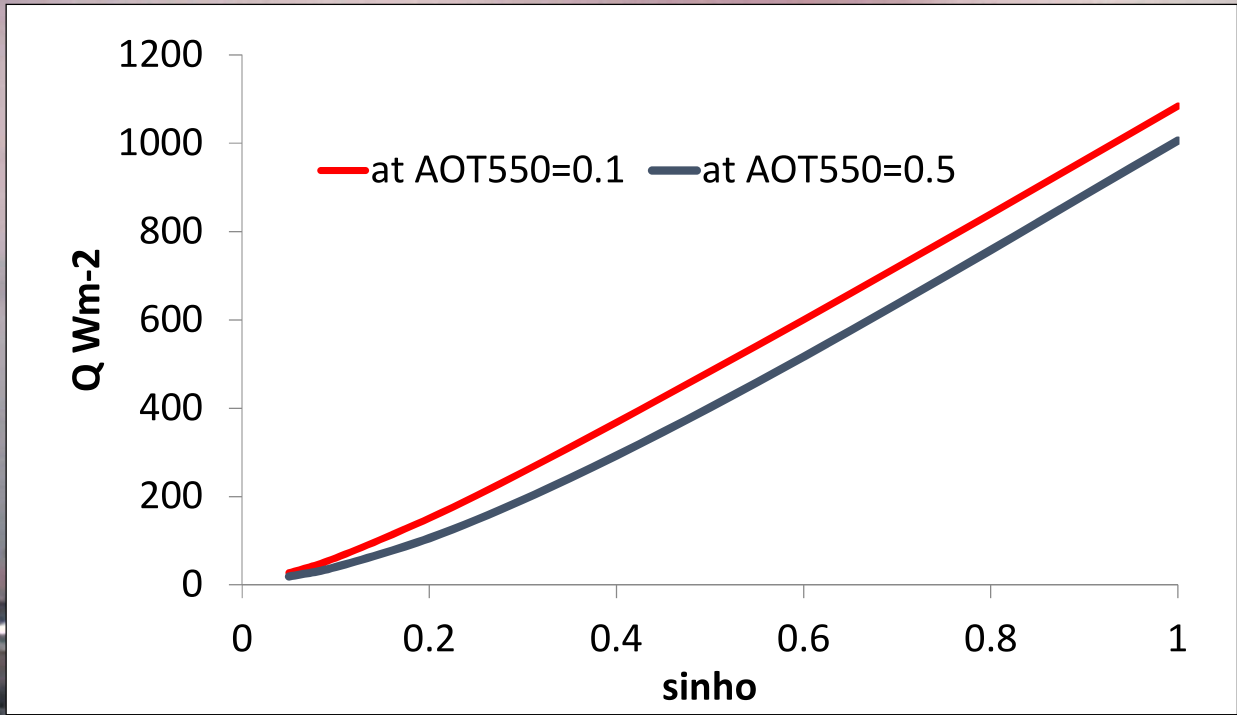
IPCC, 2013

4. Direct aerosol and cloud effect on solar irradiance

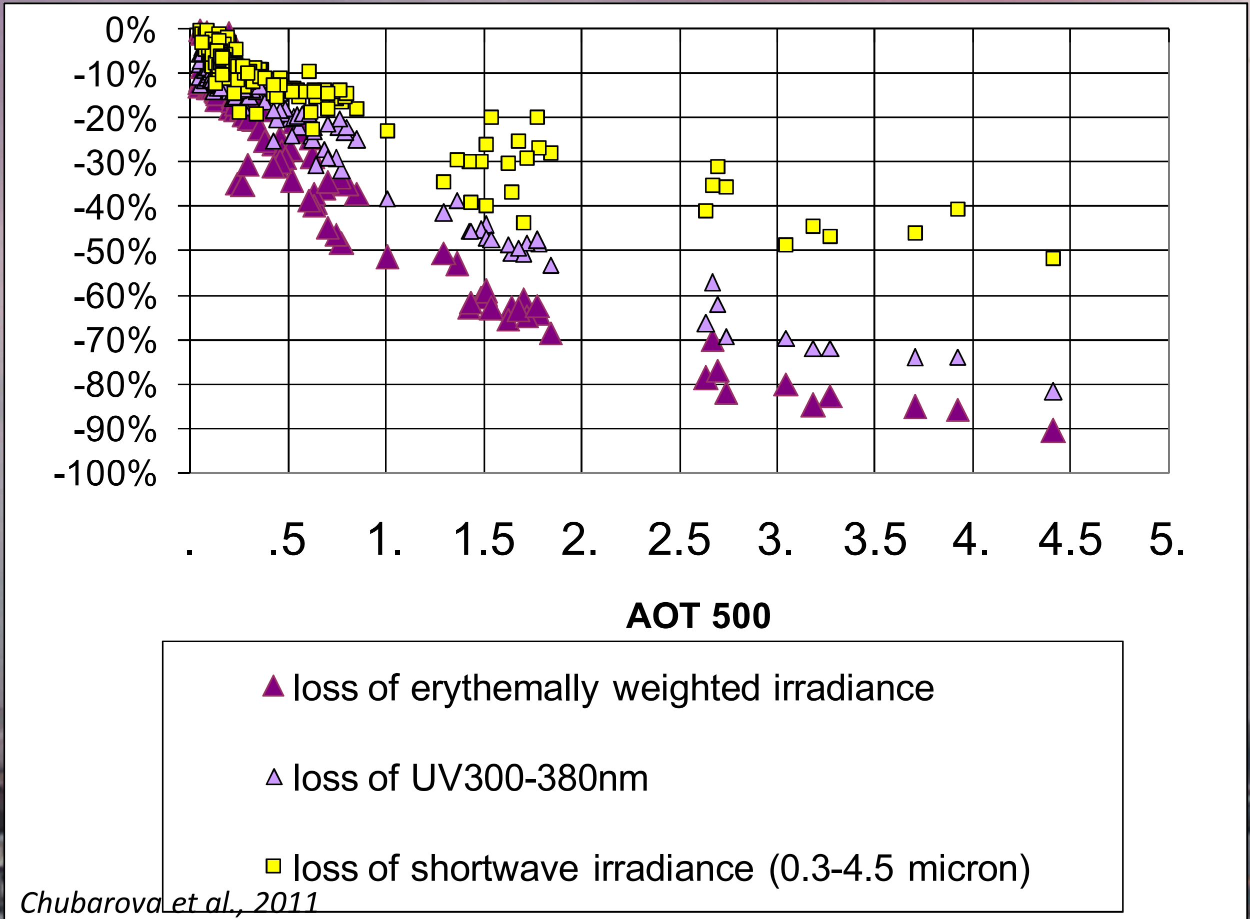
The direct effect is scattering, absorption of radiation, change in the radiation balance of the planet.



The dependence of the global shortwave radiation Q at ground vs solar height and the aerosol properties (aerosol optical thickness) of the atmosphere. Clear sky. Continental aerosol model.



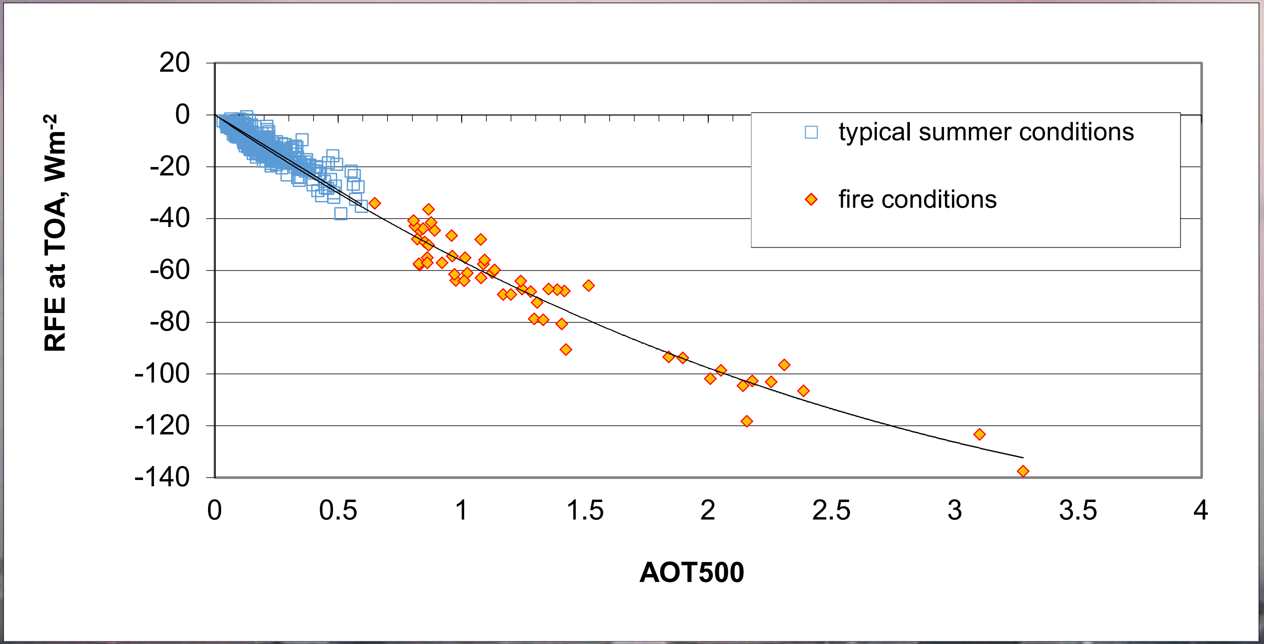
The loss in solar irradiance in different spectral regions as a function of aerosol optical thickness at 500nm Moscow. 2010.



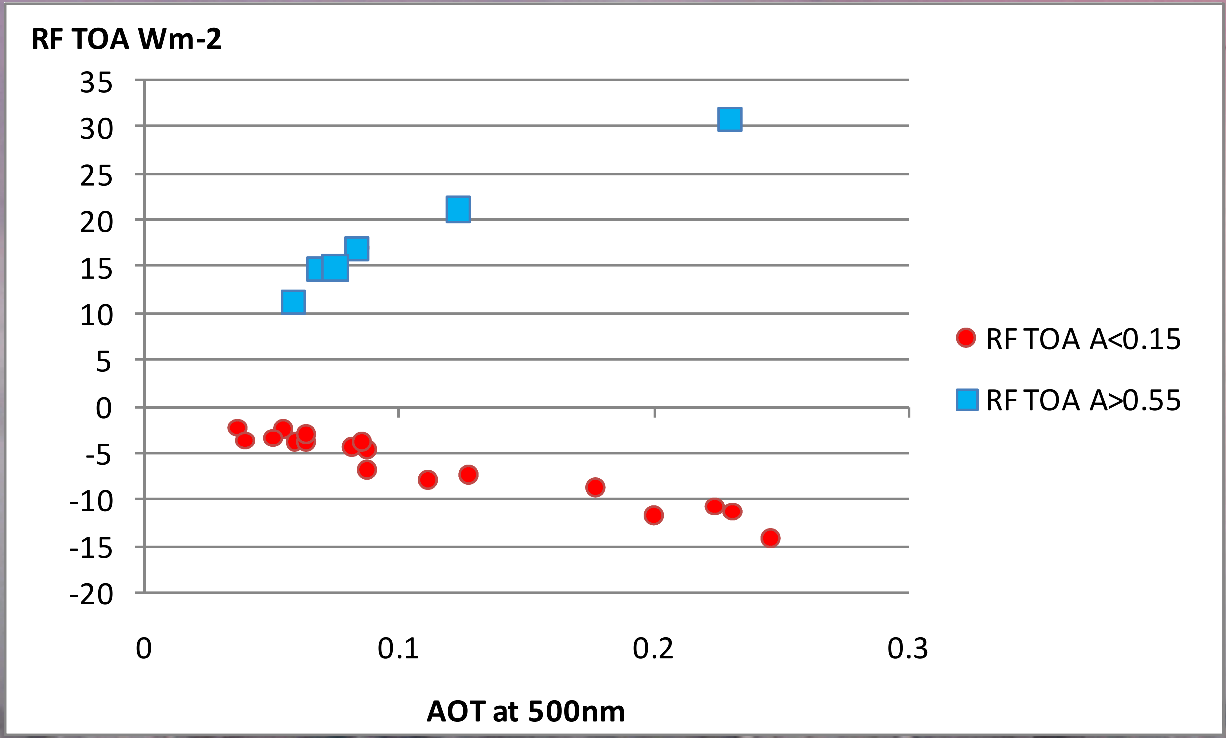
Chubarova et al., 2011

Radiative effects of aerosol at the Top of the Atmosphere (TOA) in typical and fire conditions.

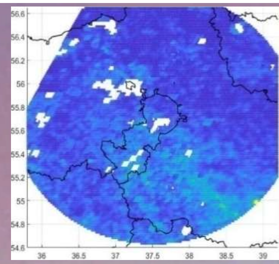
$$RFE(TOA)=F_{net,aerosol}-F_{net,no\ aerosol}$$



Depending on surface type radiative effect can be even of different sign!



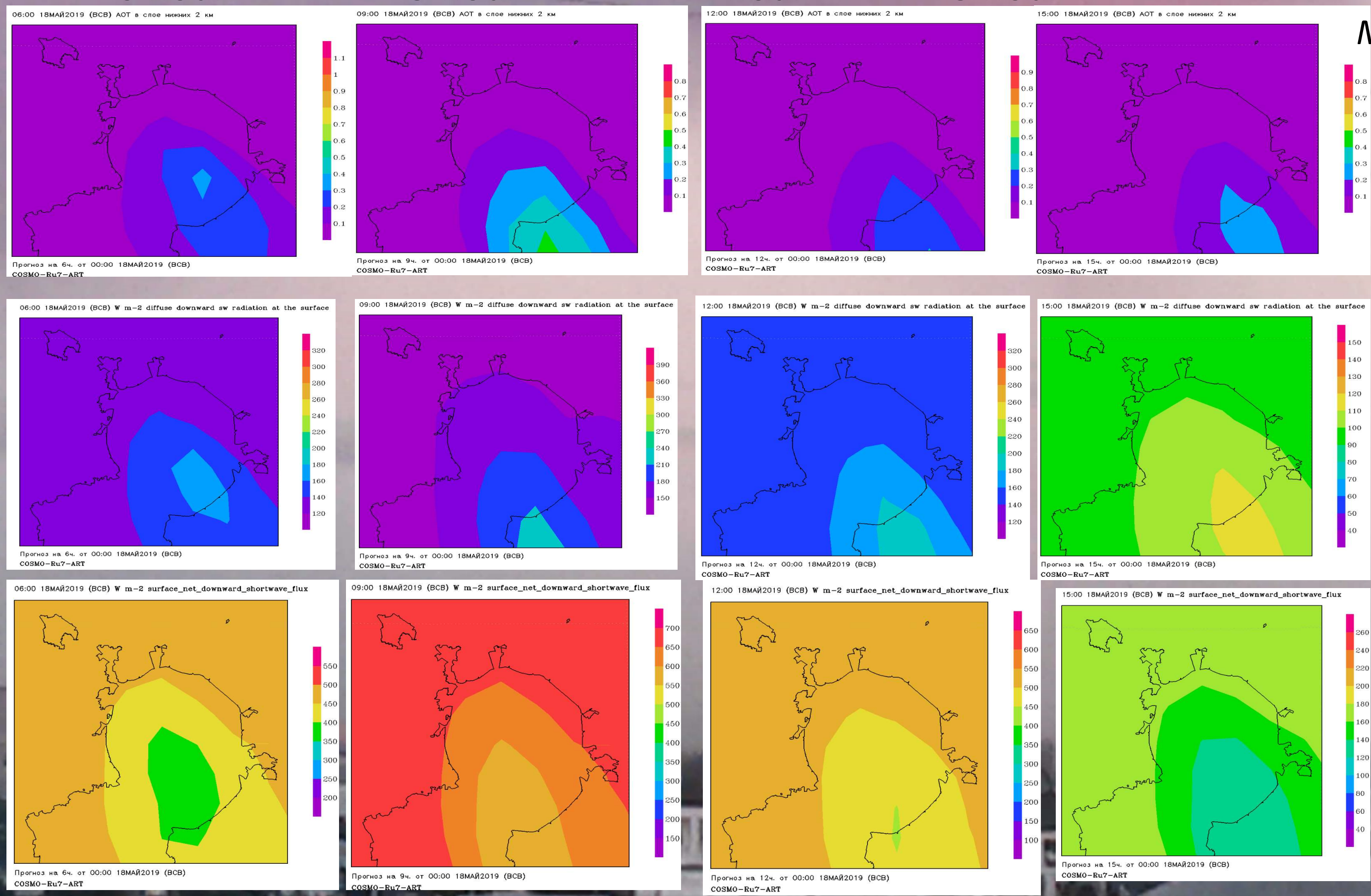
The dynamic of urban aerosol development in Moscow region and its influence on radiative characteristics of the atmosphere according to COSMO-Ru-ART. 18.05.2019.



AOT MODIS /MAIAC
May 18 2019.

6 hour 9 hour 12 hour 15 hour

wind direction



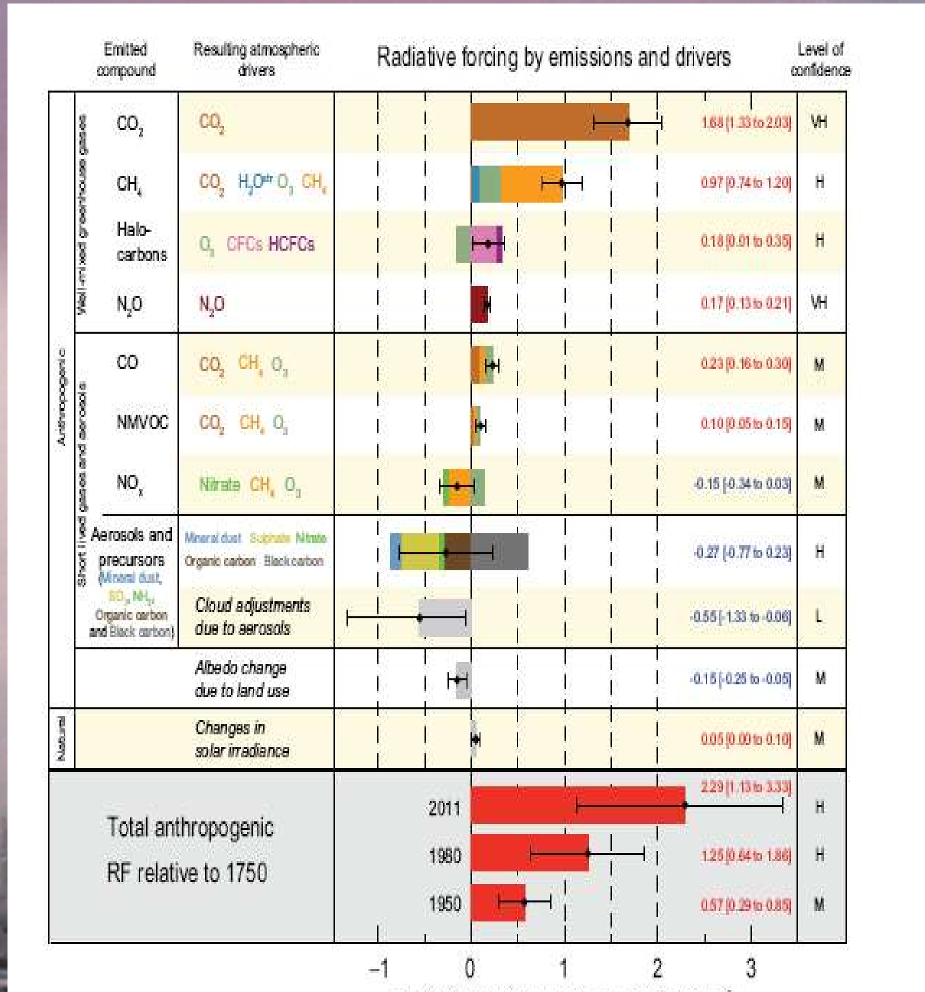
aerosol optical thickness

diffuse radiation

net radiation at ground

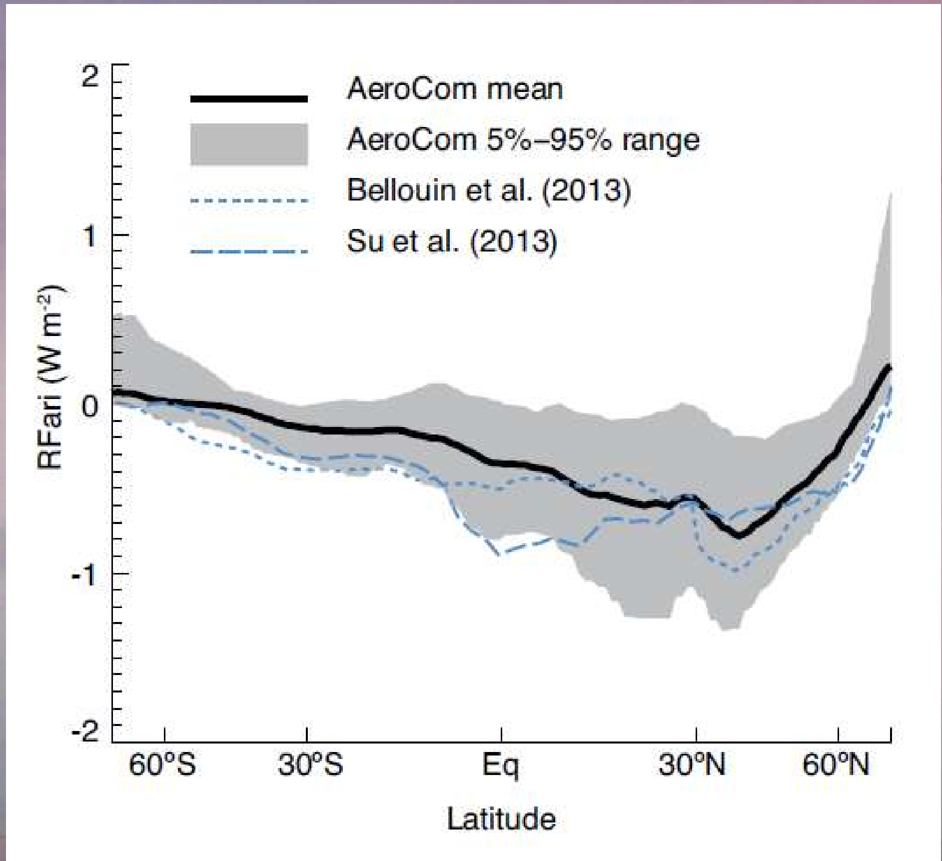
Effective Radiative forcing

- ERF(i)=Fnet(i)-Fnet(i)_at 1750
- for i-component



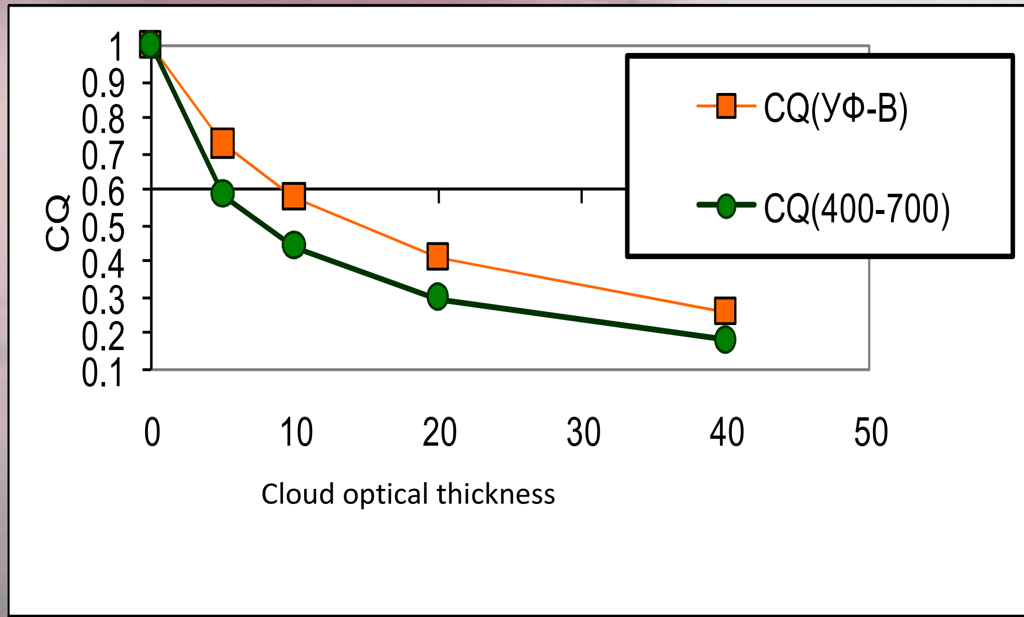
IPCC2013

Rfari (direct radiative forcing) effect of anthropogenic aerosols

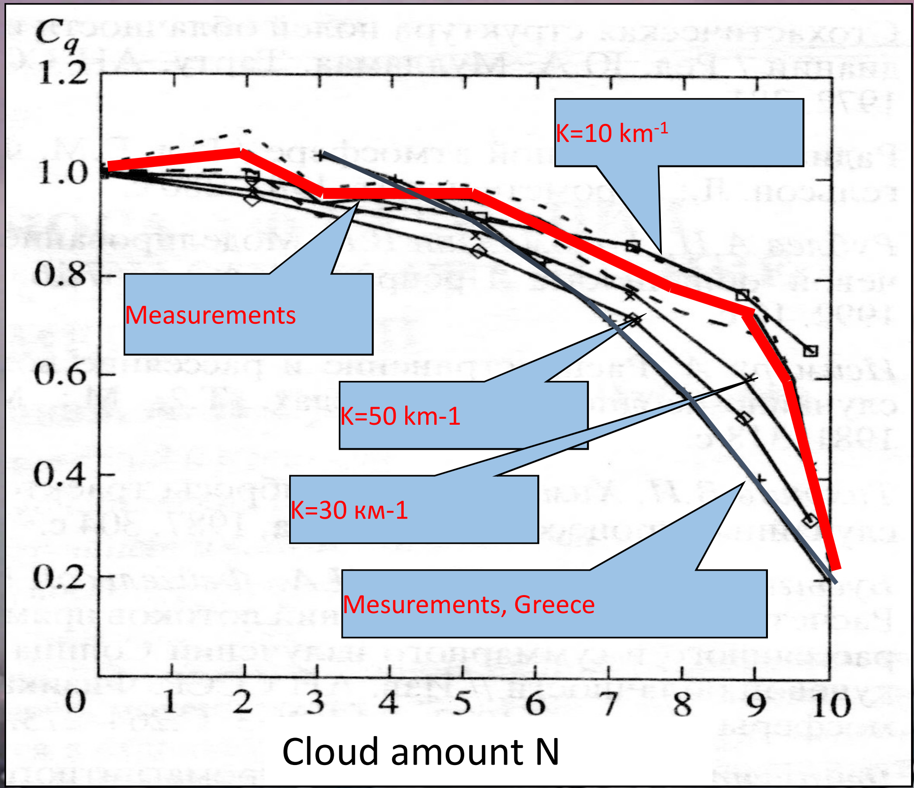


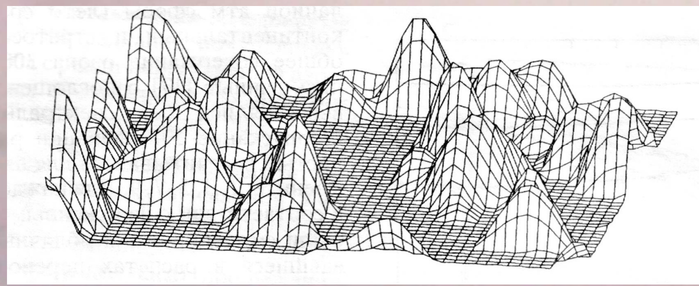
CLOUDS

The effect of overcast cloud cover on solar radiation Q transmission CQ=Q/Q0



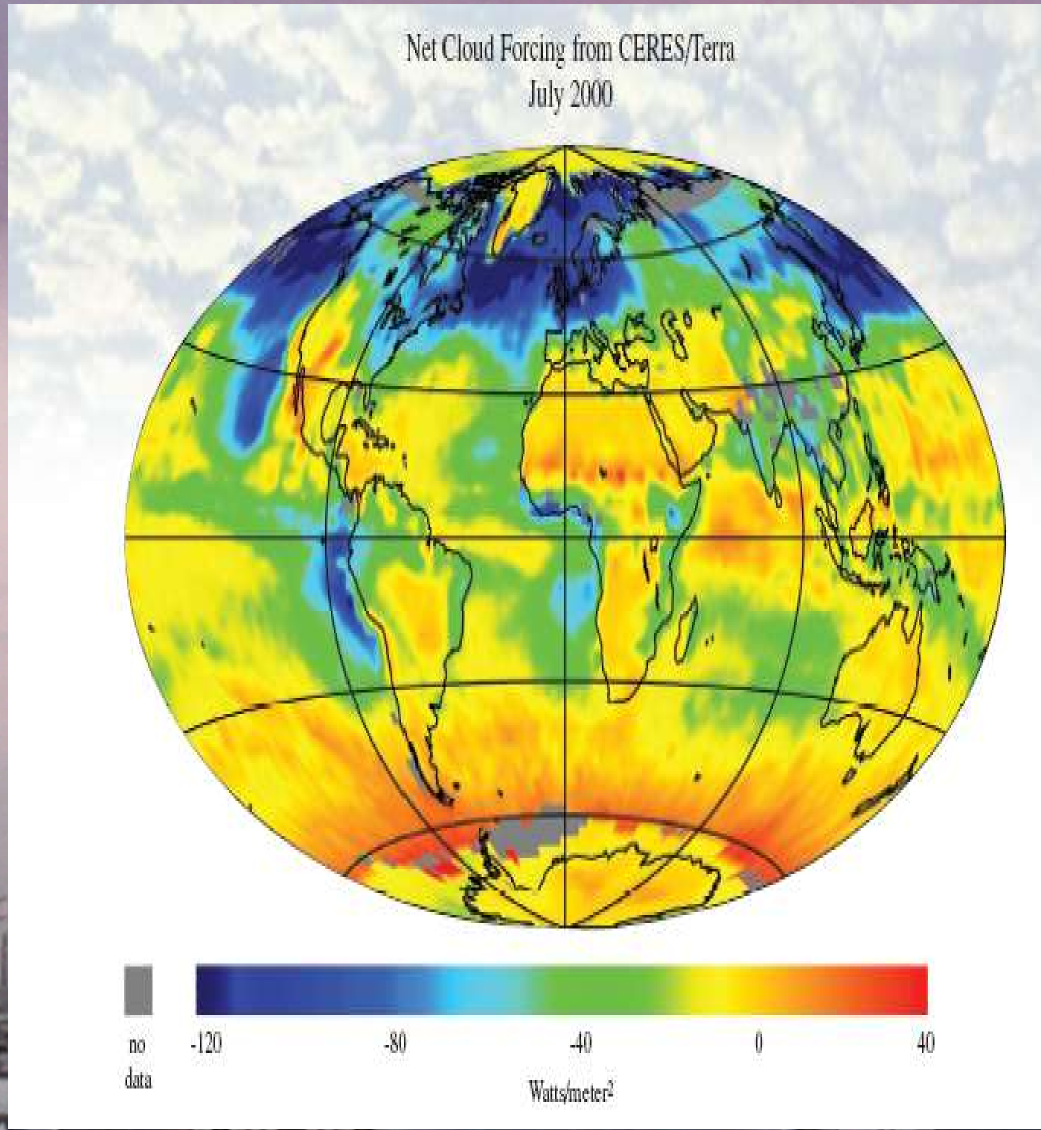
The effect of the convective cloud amount N on the solar radiation transmission according to measurement and modeling data



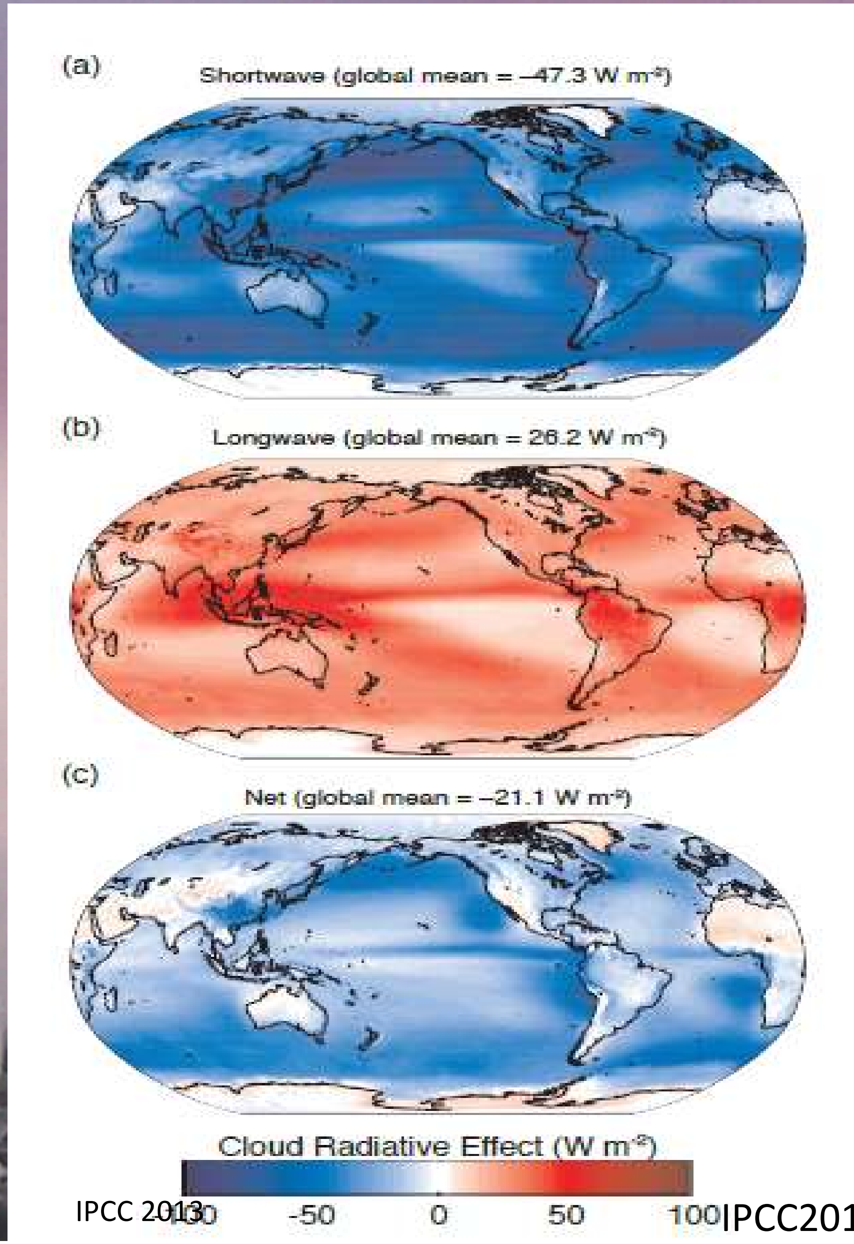


Example of cloud radiative effect at the top of the atmosphere

Cloud Radiative Effect =
=Net Radiation (cloud)-Net radiation (no cloud)



Average cloud radiative effect

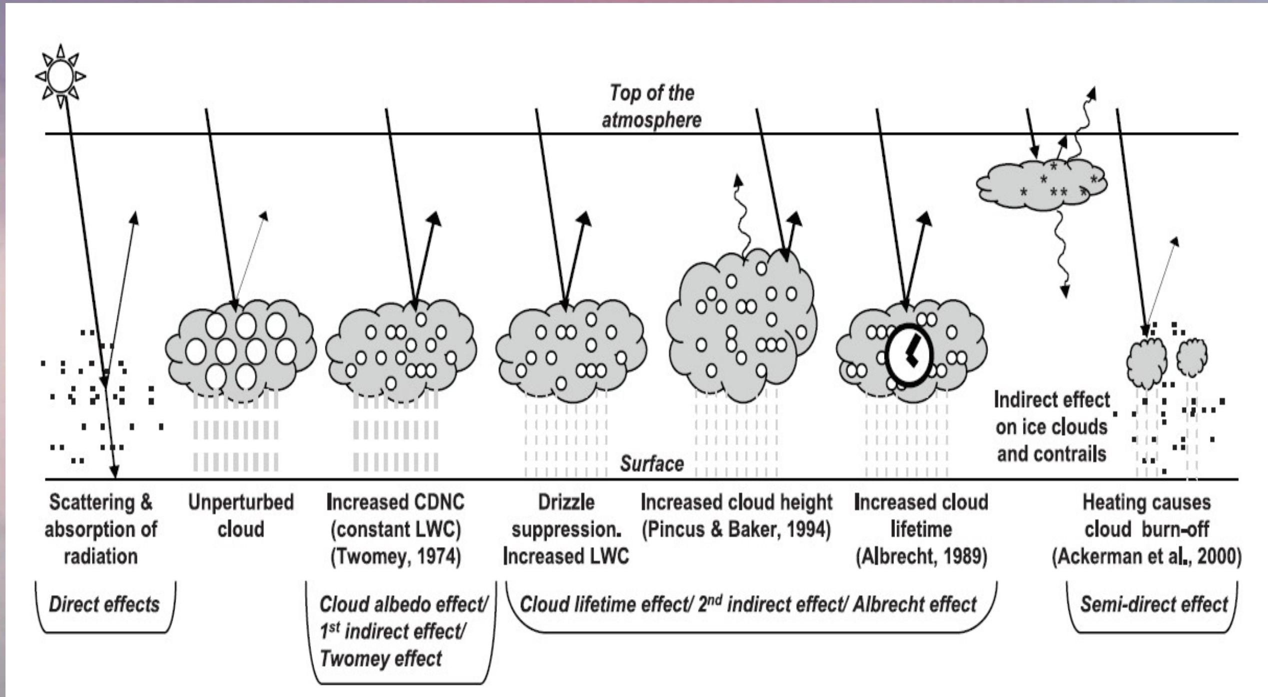


IPCC 2013

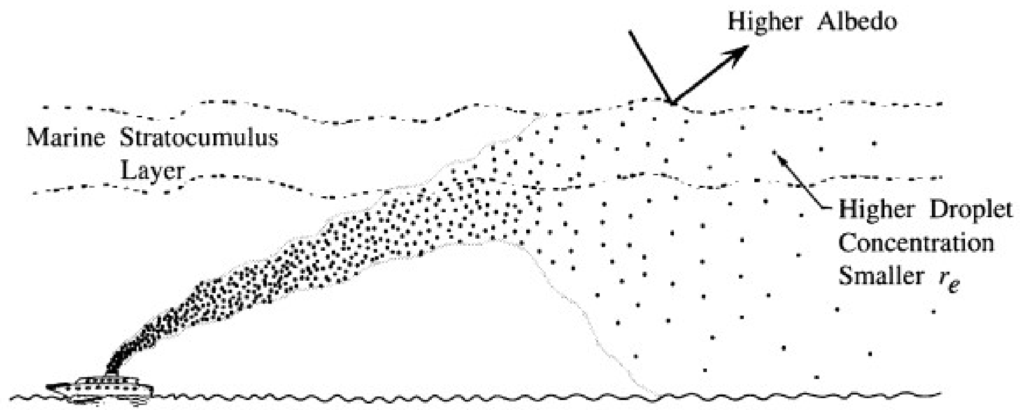
IPCC2013

Indirect cloud and aerosol effects on solar irradiance

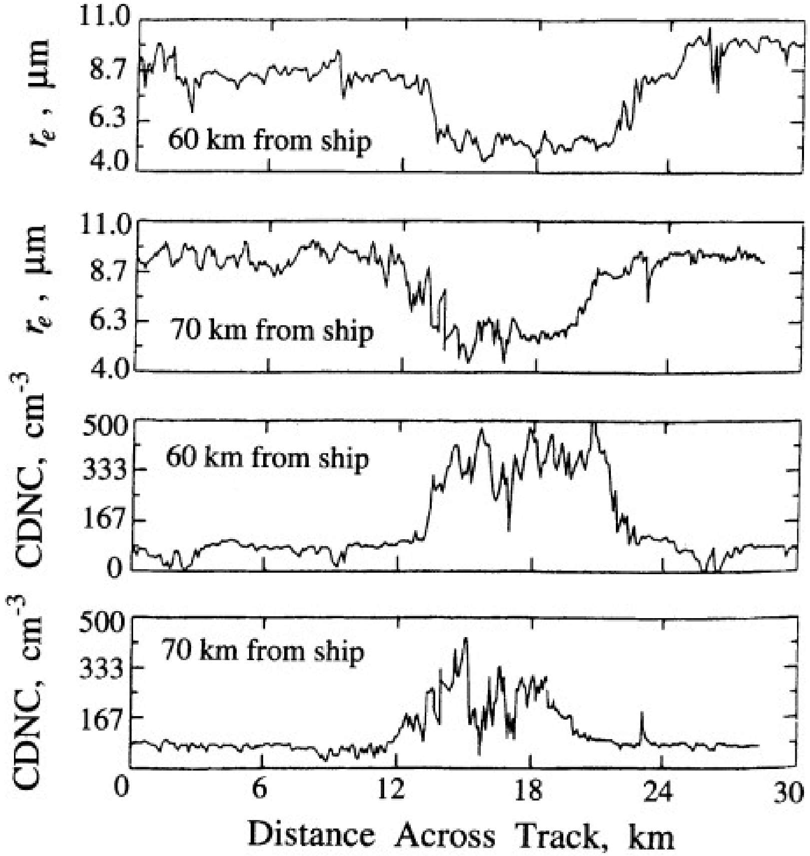
- **Indirect aerosol effects:**
- The first indirect effect (with constant water content):
- an increase in cloud albedo due to aerosol condensation nuclei - Twomey effect 1977
- The second indirect effect (when the water content in the cloud changes):
- Increasing the life of the cloud (Albrecht effect, 1989)
- Increasing the height of the cloud
- Suppression of drizzle and increase in water content. couple.



- **<u>Mixed effect (semi direct)</u>**
- Possible rad. heating of the atmosphere due to the absorbing aerosol, and a change in the relative humidity of the air due to this, which can lead to evaporation of the cloud and a change in its properties.



(b)



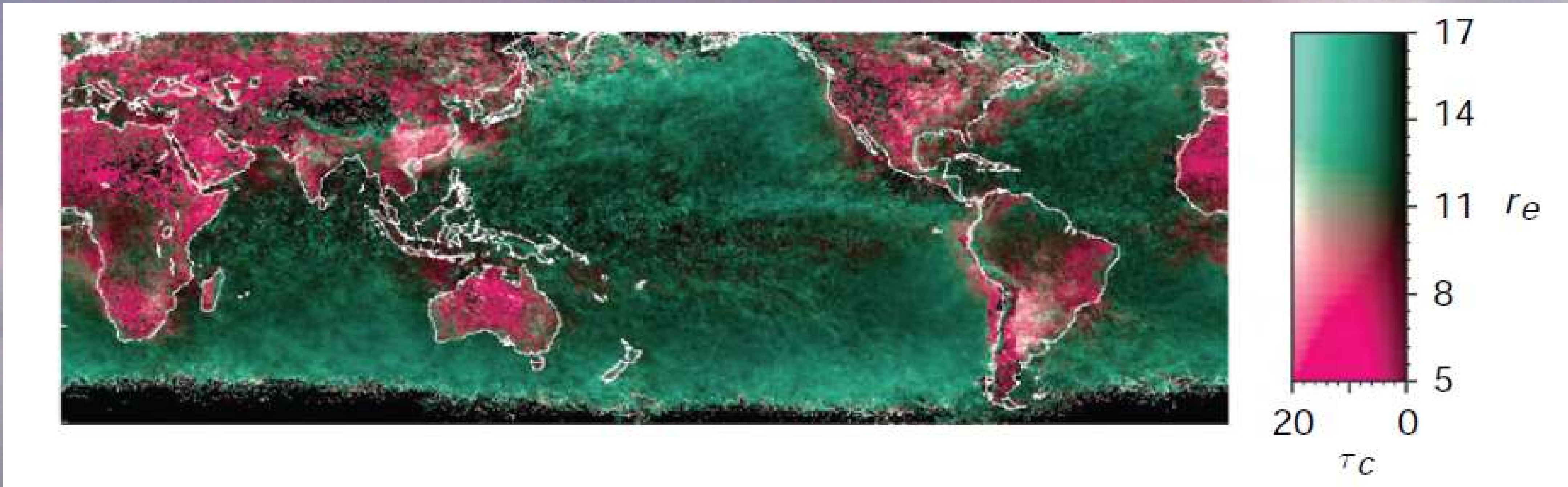
(c)

From Seinfield Pandis, 2016

FIGURE 24.16 Ship tracks: (a) satellite image at 3.7 μm wavelength of ship tracks off the western coast of the United States (courtesy of P. A. Durkee); (b) schematic of processes leading to ship tracks in marine stratocumulus clouds; (c) cloud droplet number concentration (CDNC) and effective droplet radius (r_e) measured during two transects through a ship track in cloud 60 and 70 km from the ship. The center of the ship track is at ~16 km along the transect. (Reprinted from Johnson, D. W., et al., The effects of a localised aerosol perturbation on the microphysics of a stratocumulus cloud layer, in *Nucleation and Atmospheric Aerosols 1996*, M. Kulmala and P. E. Wagner (eds.), p. 864, 1996, with kind permission from Elsevier Science Ltd. The Boulevard, Langford Lane, Kidlington OX5 1GB, UK.)

From Seinfield Pandis, 2016

Retrievals from a satellite of cloud optical thickness (${}_c$) and cloud particle effective radius (r_e in m) for low-level water clouds.



T. Nakajima et al., *"A possible correlation between satellite-derived cloud and aerosol microphysical parameters,"*
Geophys. Res. Lett. ***28***, *1172 (2001).*

Different assessment of ERFari+aci for anthropogenic aerosols

Modelling Group	Model Name	ERFari+aci from All Anthropogenic Aerosols	ERFari+aci from Sulphate Aerosols Only
CCCma	CanESM2	−0.87	−0.90
CSIRO-QCCCE	CSIRO-Mk3-6-0[b]	−1.41	−1.10
GFDL	GFDL-AM3	−1.60 (−1.44[a])	−1.62
GISS	GISS-E2-R[b]	−1.10[a]	−0.61
GISS	GISS-E2-R-TOMAS[b]	−0.76[a]	
IPSL	IPSL-CM5A-LR	−0.72	−0.71
LASG-IAP	FGOALS-s2[c]	−0.38	−0.34
MIROC	MIROC-CHEM[b]	−1.24[a]	
MIROC	MIROC5	−1.28	−1.05
MOHC	HadGEM2-A	−1.22	−1.16
MRI	MRI-CGM3	−1.10	−0.48
NCAR	NCAR-CAM5.1[b]	−1.44[a]	
NCC	NorESM1-M	−0.99	
Ensemble mean		−1.08	
Standard deviation		+0.32	

Global average direct and indirect anthropogenic effects – effective radiative forcing:

ERFari – (aerosol–radiation interactions)
direct effect
-0.27 Wm-2

ERFaci (aerosol-cloud interaction)
non direct effect
-0.96 Wm-2

Total
-1.23 Wm-2
2014 relative to 1850

Compared with appr. 2.69 Wm-2 for CO2

IPCC 2021

IPCC 2021

6. The example of indirect effects during lockdown COVID-19 in Moscow megacity due to aerosol-cloud interaction.

Moscow megacity:
Area – 2561.5 sq.km
Population - 12.5 million (live in the city permanently)
Personal car fleet - 4.7 million vehicles (7.7 million with Moscow region)
3.6 million cars are moving around Moscow every day

Khlestova et al., 2021

typical situation on the streets during rush hour

streets during lockdown period

Methods for retrieval the concentration of cloud condensation nuclei (N_{CCN})

1 method

$$N_d = c_1 R_{eff}^{-5/2} COT^{1/2}$$

(Quaas et al., 2006)

2 method

$$N_d = c_2 LWP^{-5/2} COT^3$$

(McComiskey et al., 2009)

N_d – Number concentration of liquid cloud particles, m^{-3}
R_{eff} – Effective radius of liquid cloud particles, m
COT – Cloud optical thickness
LWP – Liquid water path, kg/m^2

Methods assumptions:

- Liquid (warm) cloud only
- Gamma size-distribution function
- Sub adiabatic cloud
- $N_d|_z = const$
- $N_{CCN} \approx N_d$ at the cloud base

Conditions:

- Sun elevation 25° at least
- Sensor zenith less than 45°
- Only liquid cloud phase (based on MODIS data)
- 1-2 cloud layers
- Cloud optical thickness more than 5

INdirect effects of aerosol.

Smaller aerosol content may affect cloud properties.

Retrievals of N_{CCN} (cm^{-3}) from satellite MODIS data.

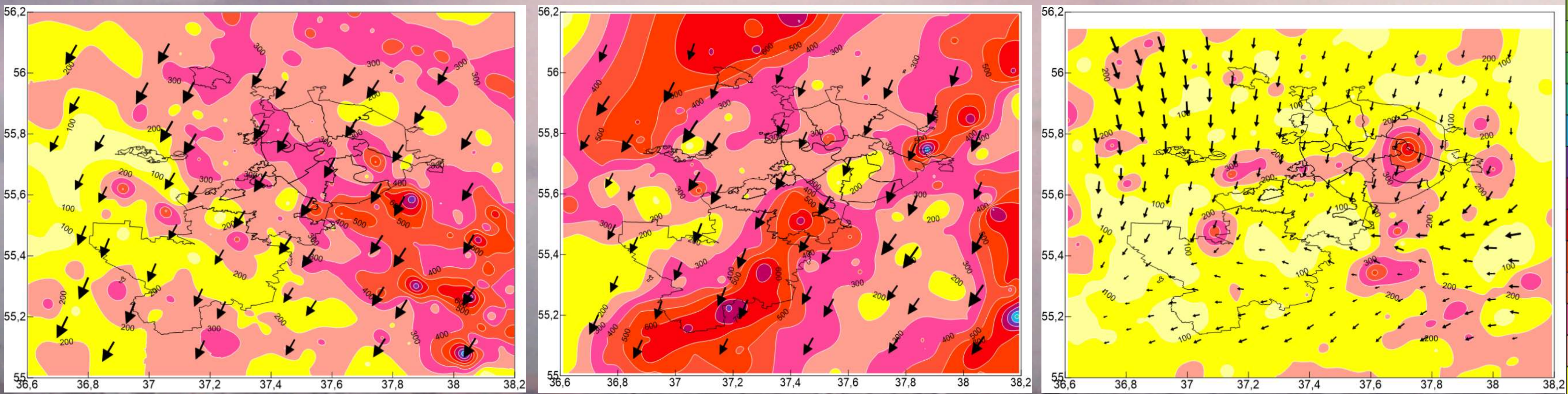
Only northern advection cases

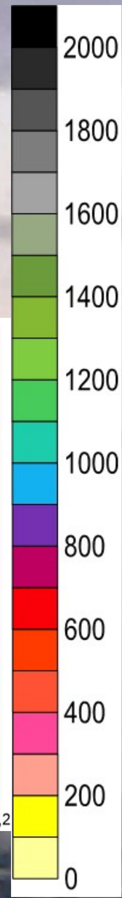
N_{CCN} – Number concentration of Cloud Condensation Nuclear

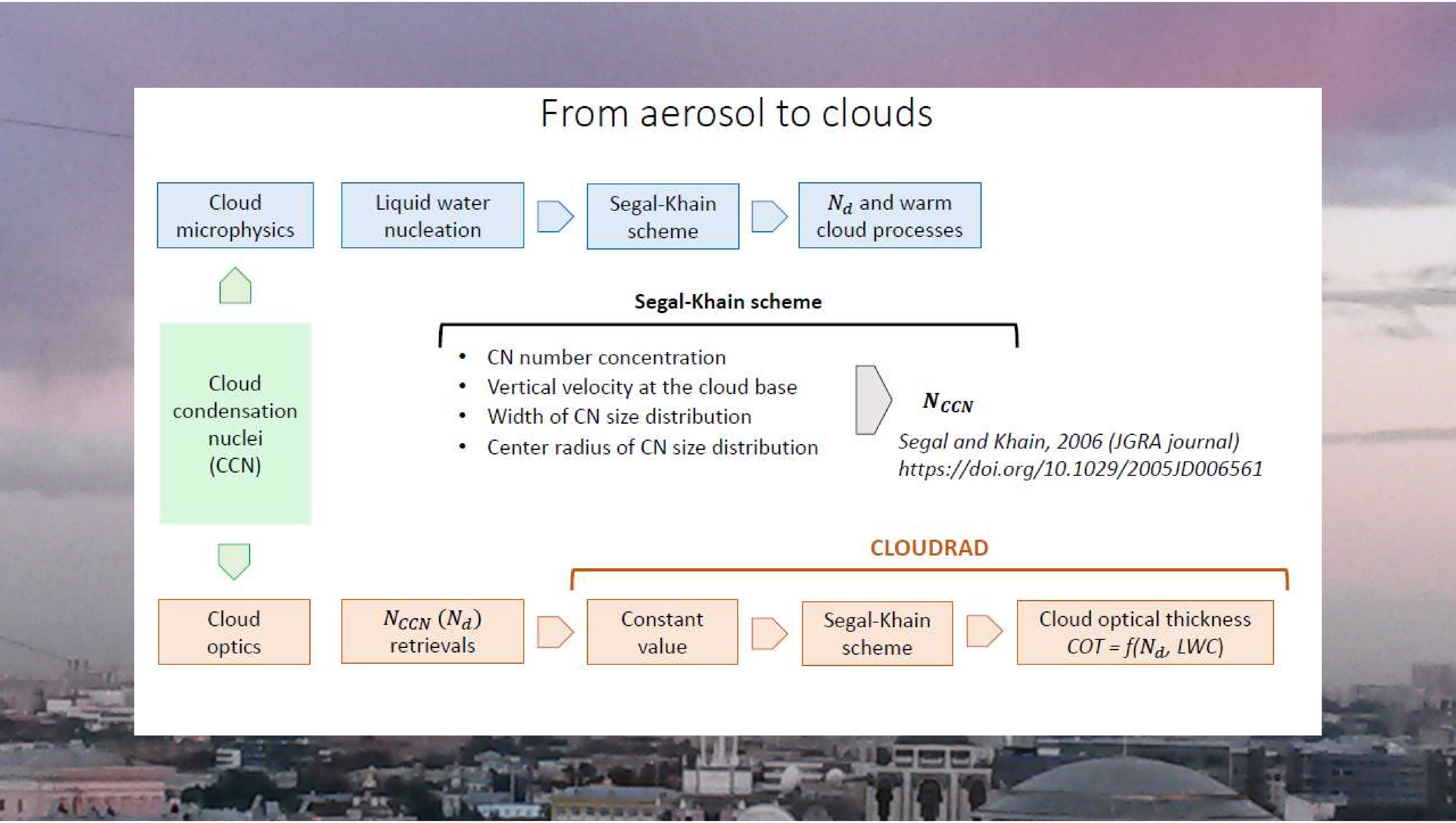
22/04/**2018** 09:05 a.m.

02/05/**2019** 10:55 a.m.

22/05/**2020** 10:45 a.m. – lockdown period



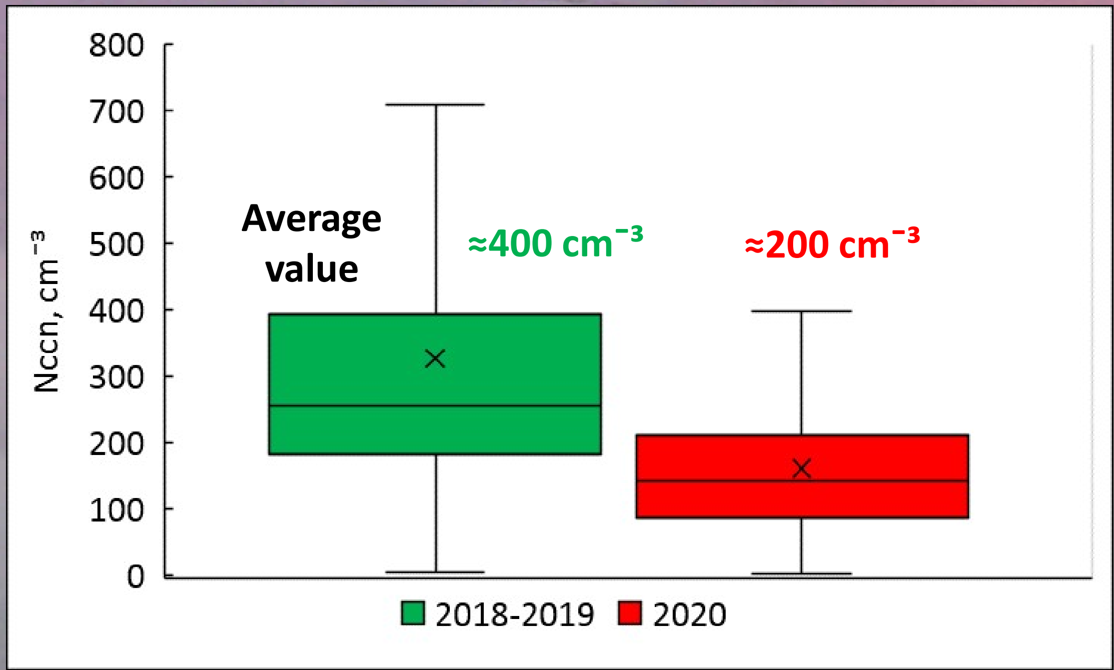



From aerosol to clouds
Cloud microphysics
Liquid water nucleation
Segal-Khain scheme
N_d and warm cloud processes
Segal-Khain scheme
• CN number concentration
• Vertical velocity at the cloud base
• Width of CN size distribution
• Center radius of CN size distribution
N_{CCN}
Segal and Khain, 2006 (JGRA journal)
https://doi.org/10.1029/2005JD006561
Cloud condensation nuclei (CCN)
CLOUDRAD
Cloud optics
N_{CCN} (N_d) retrievals
Constant value
Segal-Khain scheme
Cloud optical thickness
COT = f(N_d, LWC)

Retrievals of N_{CCN} (cm^{-3}) by 1 km MODIS satellite data

N_{CCN} – Number concentration of Cloud Condensation Nuclear

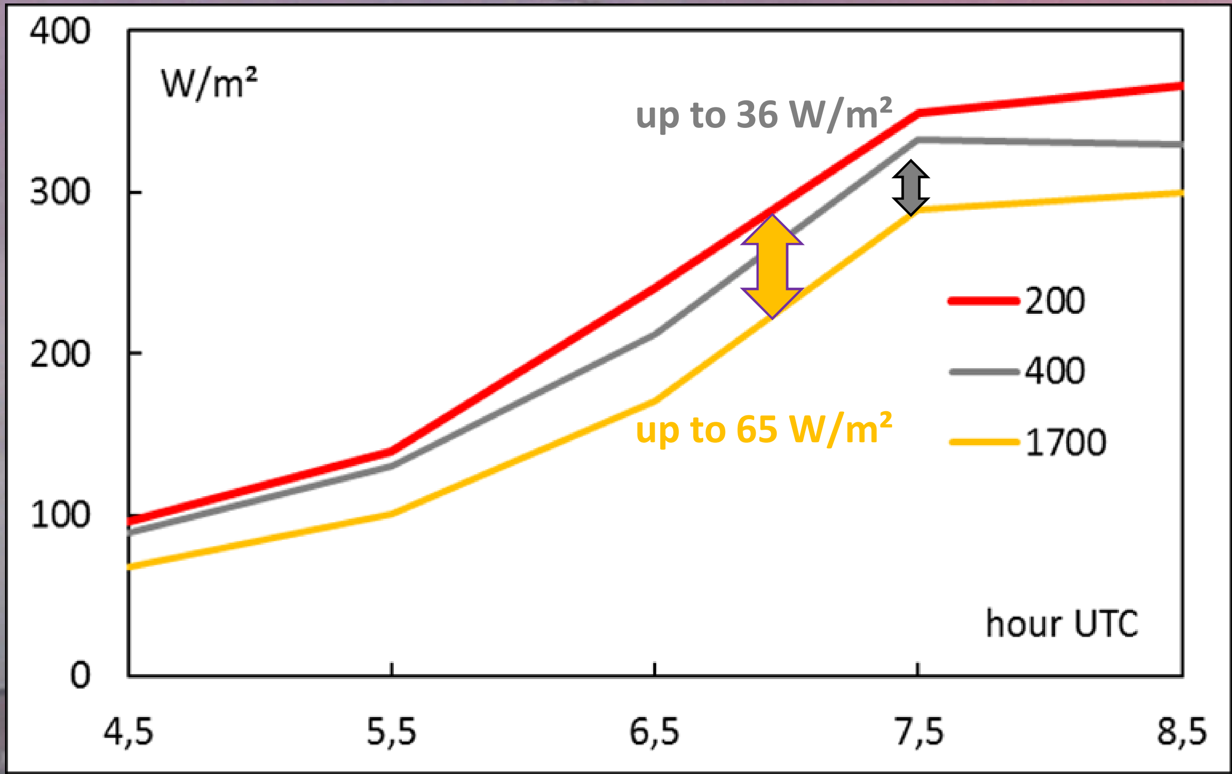
Cases of northern advection



The most important selection conditions:

- Only liquid cloud coverage
- 1-2 layers coverage

The global irradiance for different droplets R_{eff} due to different N_{CCN}



Conclusions

- Importance and yet large uncertainty of the indirect aerosol-cloud interaction Faci effects on radiation.

- Need further studies!

Questions?