

Aerosol processes, properties, variability and modelling

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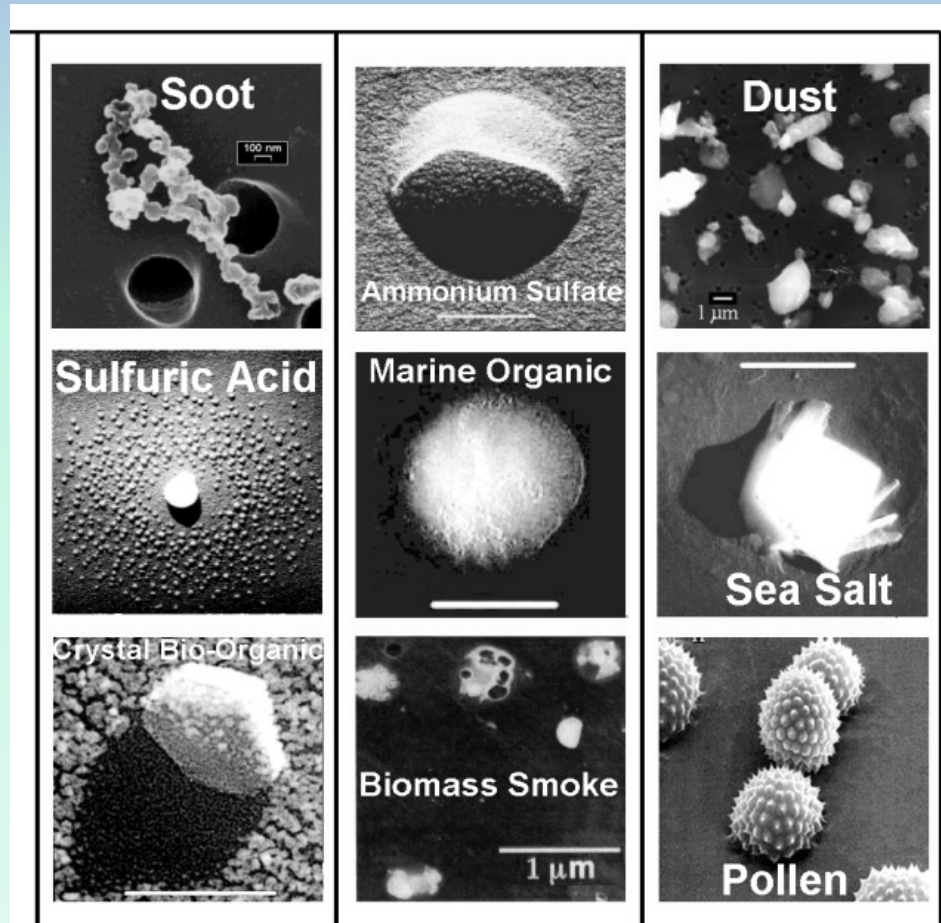
Reference books

**Introduction to atmospheric chemistry (Jacob)
Atmospheric Chemistry and Physics (Seinfeld ,Pandis)
Fundamentals of atmospheric modelling (Jacobson)**

Aerosol ?

Aerosol: dispersed or condensed phases suspended in a gas

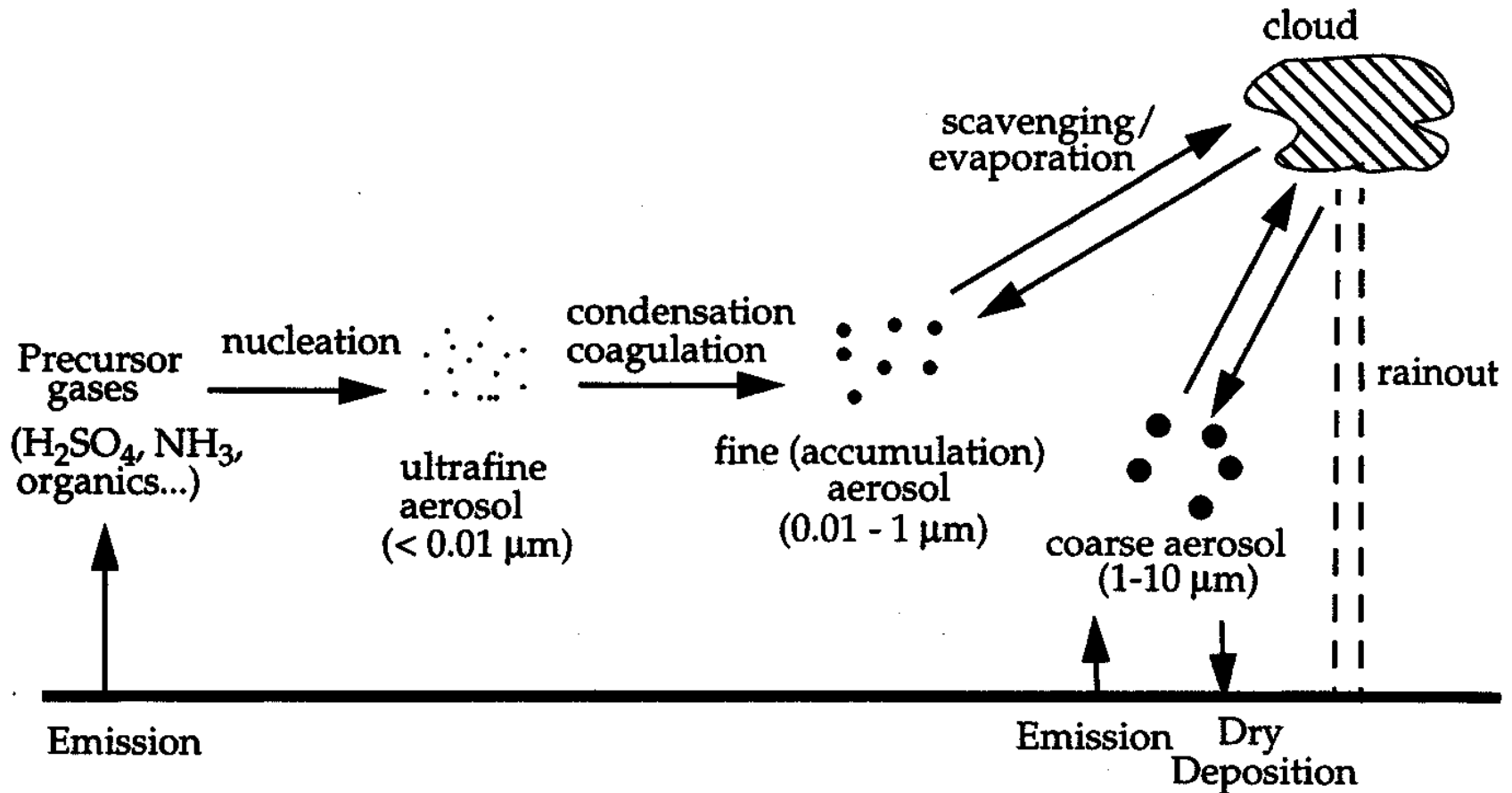
Size range: $0.001\ \mu\text{m}$ (molecular cluster) to $100\ \mu\text{m}$



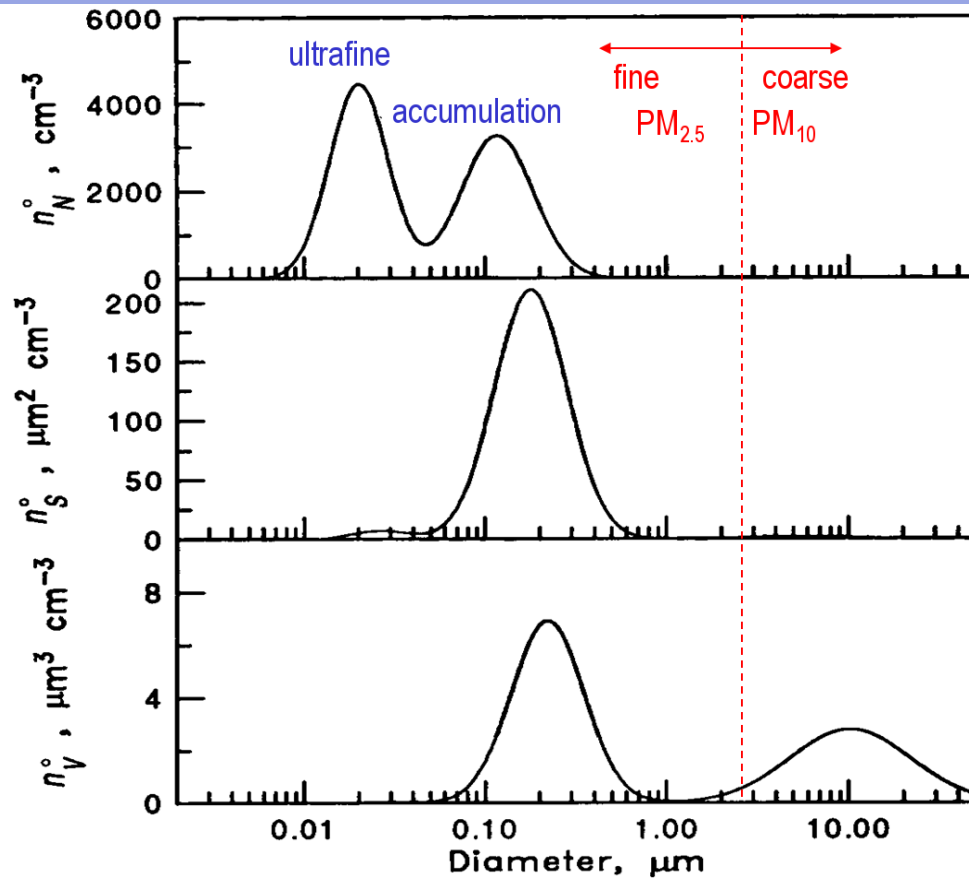
Origin of the atmospheric aerosol

Aerosol: dispersed or condensed phases suspended in a gas

Size range: $0.001\ \mu\text{m}$ (molecular cluster) to $100\ \mu\text{m}$



Aerosol size distribution



$$n(D_p) = \frac{dN}{dD_p}$$

N = number concentration (particles/ cm^3)

Number

$$N = \int_0^{\infty} n(D_p) dD_p$$

$$n_s(D_p) = \frac{dS}{dD_p}$$

$$= \pi D_p^2 n(D_p)$$

Surface

$$n_v(D_p) = \frac{dV}{dD_p}$$

$$= \frac{\pi}{6} D_p^3 n(D_p)$$

**Volume
(mass)**

It is also current to use the *particle volume* instead to of *the particle diameter* as the independent variable of the distribution:

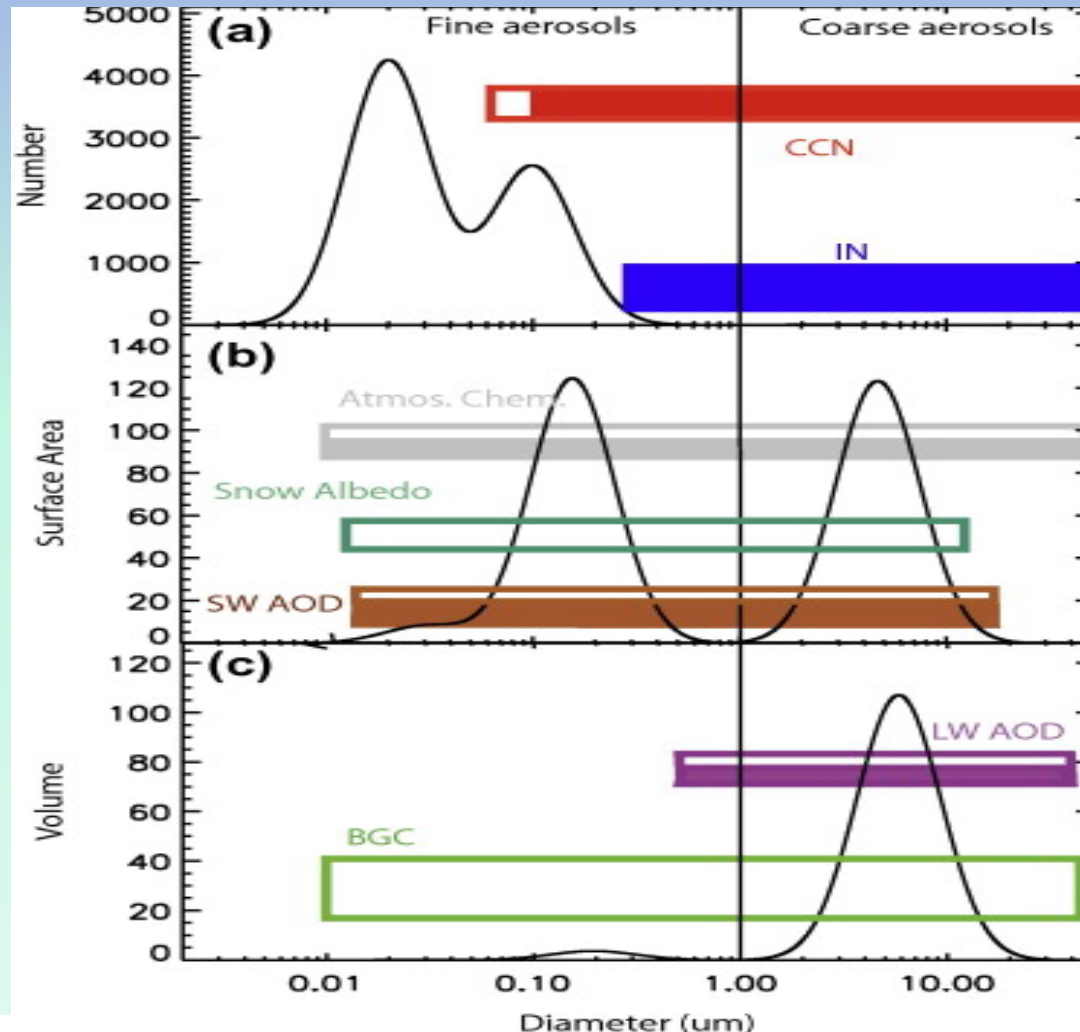
$$n(D)dD = n(v)dv \Rightarrow n(v) = \frac{\pi}{2} \cdot D^2 \cdot n(D)$$

↓

Number of particles / cm^3 having their volume between v and $v+dv$

Size vs. Earth system impacts

Size distribution and chemical properties will ultimately determine interactions with Earth system and impacts



Atmospheric chemistry modelling

$$\frac{\partial \chi_i}{\partial t} = \underbrace{-\bar{V} \cdot \nabla \chi_i + F_H + F_V + T_{CUM}}_{\text{Transport}} + \underbrace{S_{\chi_i}}_{\text{Primary Emissions}} - \underbrace{R_{w,ls} - R_{w,cum} - D_{dep}}_{\text{Removal terms}} + \underbrace{\sum \mathcal{Q}_{p_i} - \mathcal{Q}_{l_i}}_{\text{Physico-chemical transformations}}$$

For aerosols

Bulk schemes : use the above equation for each aerosol species, $Q_p - Q_l$ accounts implicitly for nucleation , condensation / evaporation. Assume a typical size distribution when needed. Numerically efficient.

Size resolved schemes: work explicitly with the size distribution evolution

The equivalent of chemistry operator is:

$$\frac{\partial n_i(v, t)}{\partial t} = \left[\frac{\partial n_i(v, t)}{\partial t} \right]_{nuc} + \left[\frac{\partial n_i(v, t)}{\partial t} \right]_{cond / eva} + \left[\frac{\partial n_i(v, t)}{\partial t} \right]_{coag}$$

Nucleation

Formation of a new phase

In the atmosphere: vapour \rightarrow liquid, vapour \rightarrow solid

In 1897 first evidences of new particle formation in the atmosphere, but only around 2000 instrumentation to quantitatively measure it appeared

Types of nucleation

- **Homogeneous nucleation: No foreign nuclei or surfaces**
- **Heterogeneous nucleation: Nucleation on a foreign substance**
- **Ion induced nucleation: Nucleation on charged particles**

Number of species involved

- **One: homomolecular nucleation**
- **Two or more: heteromolecular or binary, ternary, ...**

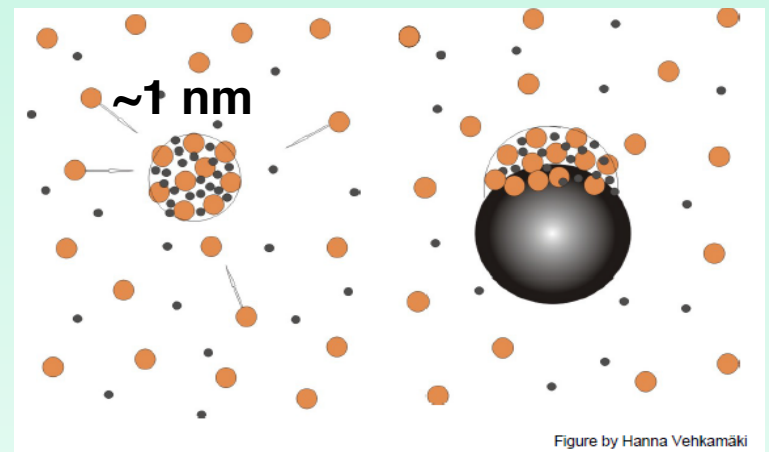


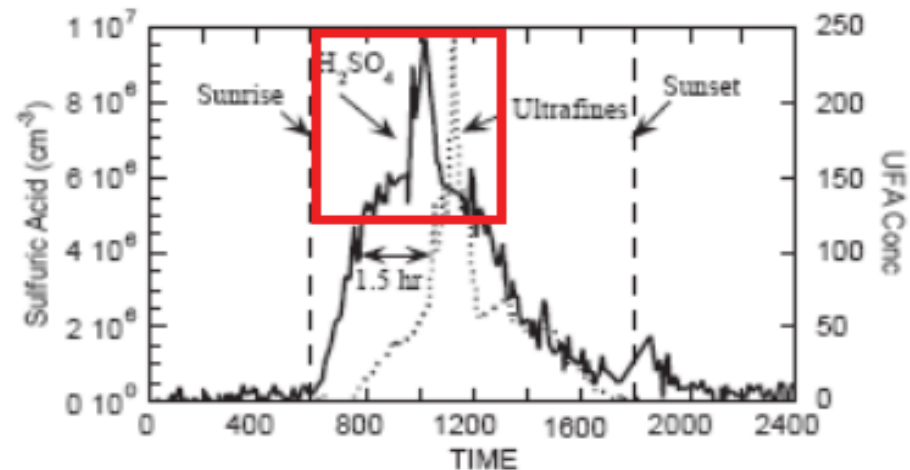
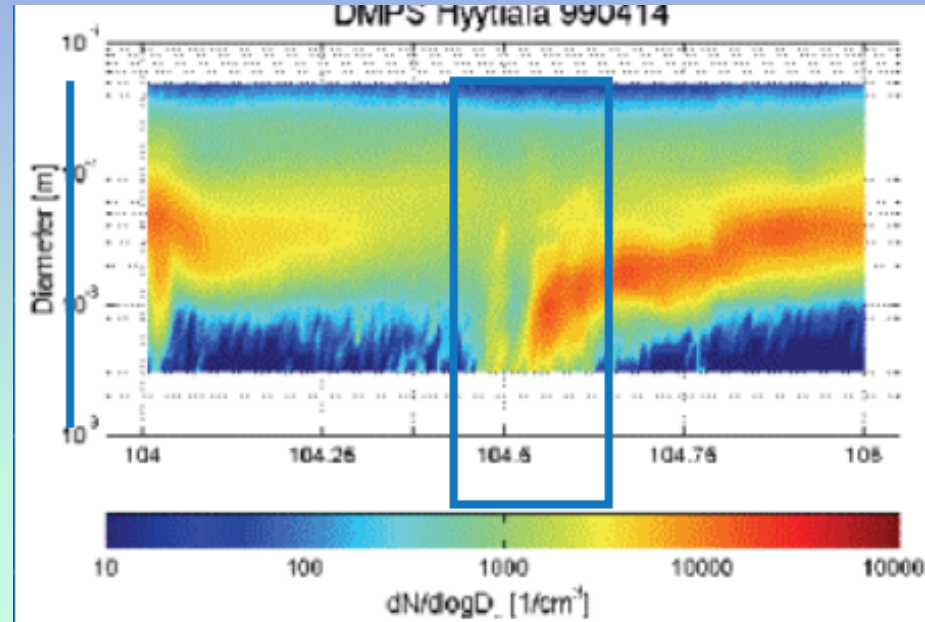
Figure by Hanna Vehkamäki

Potential aerosol nucleation pathways in the atmosphere

- Binary water-sulphuric acid nucleation (mainly free troposphere)
- Ternary water-sulphuric acid-ammonia nucleation (lower troposphere)
- Ion-induced nucleation (higher troposphere more likely / role of cosmic rays)
- Nucleation of some organic compounds, ion induced (Kirkby et al, nature, 2016)
- High uncertainties remaining notably due to the scale of observation
- Active field e.g. <http://cloud.web.cern.ch/cloud>

Particle formation takes place ...

- During daytime, suggesting that photochemistry plays an important role
- In presence of only a few pre-existent particles
- In presence of a large source of precursor vapor, of photochemical or of biogenic origin
- Sometimes at low temperatures



Classical nucleation theory

Gibbs Free energy

Variation of internal energy in a open system (or of variable composition)

$$dU = \underset{\substack{\uparrow \\ \text{Mechanical work produced}}}{-P.dV} + \underset{\substack{\uparrow \\ \text{Variation of energy linked to} \\ \text{the micro-state change of} \\ \text{the system}}}{T.dS} + dG \rightarrow \text{Energy variation not tied to mechanical work and entropy variation: e.g chem reaction, electrical work, osmotic phenomena, phase transition, creation of an interface ...}$$

For given T,P conditions

Spontaneous processes : $dG < 0$

Thermo eq. : $dG = 0$

Chemical potential = amount of free energy linked to the variation of the amount (n_i) of one component of the system (e.g as a result of a chemical reaction or change of phase)

$$\mu_i = \left(\frac{\partial G}{\partial n_i} \right)_{T,p,n_j}$$

$$dG = \sum_{i=1}^k \mu_i dn_i$$

Equilibrium for phase change (e.g vapor/liquid) of a given compound

$$\mu_1(v) = \mu_2(l)$$

Consider the homogeneous formation of a H2SO4 cluster (of radius a) from its vapor : $(1)(v) \rightarrow (2)(l)$

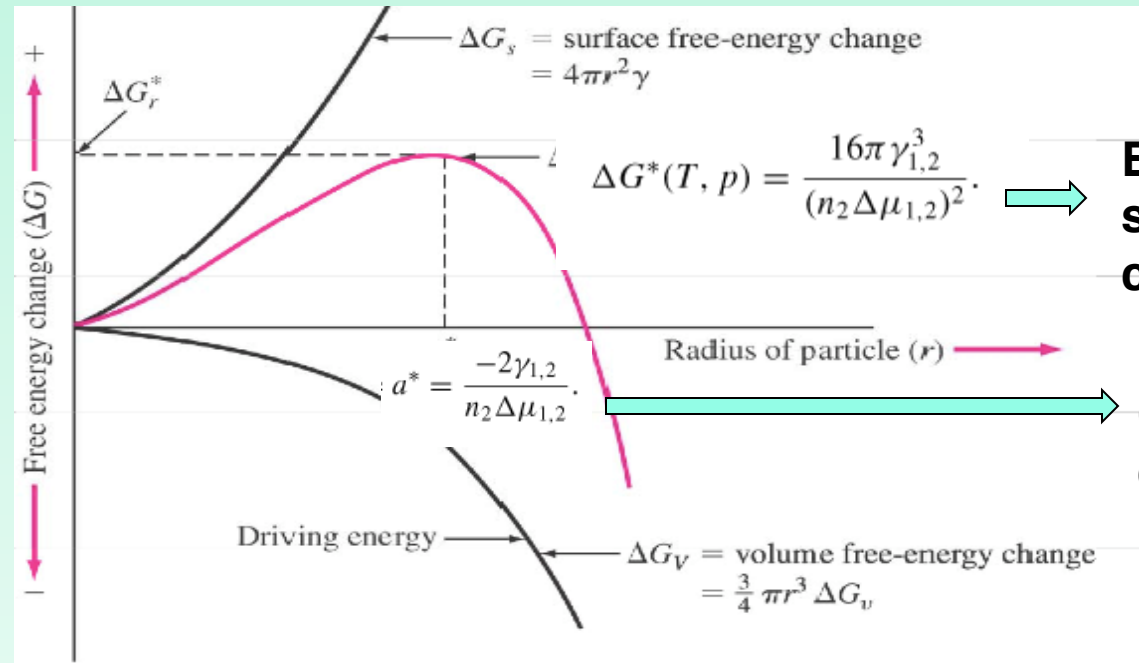
Free energy variation for this transformation :

$$\Delta G(a) = n_2 \frac{4\pi a^3}{3} (\Delta\mu_{1,2}) + 4\pi a^2 \gamma_{1,2},$$

Number of mol. entering the cluster.
 n_2 = mol. density of the condensed phase

$$\mu_{2(l)} - \mu_{1(v)} = -k.T.\ln\left(\frac{P_i}{P_{i,s}^{eq}}\right) < 0 \text{ if s.sat}$$

Free energy increase as a result of spherical surface formation > 0 .
 γ : Surface tension for h2so4 (J/m2)



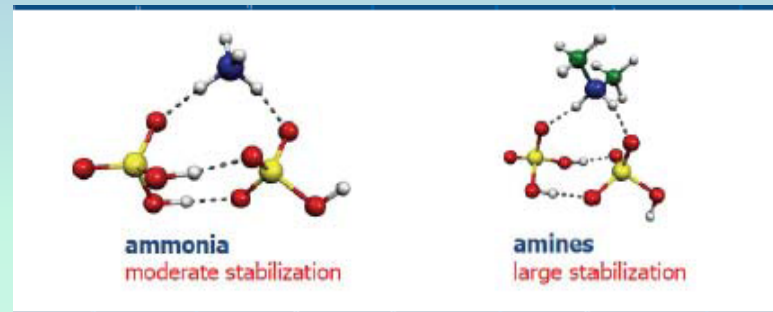
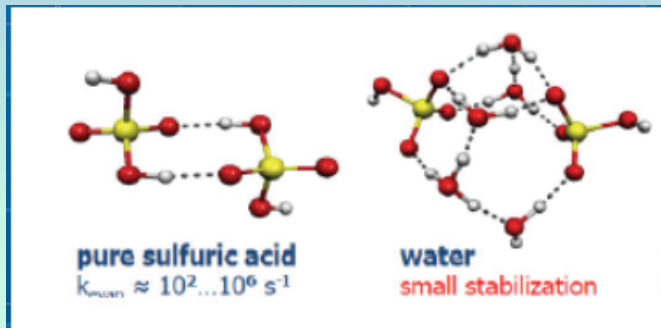
Energy barrier for the spontaneous formation of critical cluster

Critical radius for stable cluster to form

Why are heteromolecular homogeneous processes more likely ...

Other compounds can lower the **activity** (and so the chemical potential) of species i in solution (eg H₂SO₄), while the gas phase chemical potential is not affected :

$\Delta\mu_{1,2}$ increase , a^* and the energy barrier decrease



Why are heterogeneous processes more likely ...

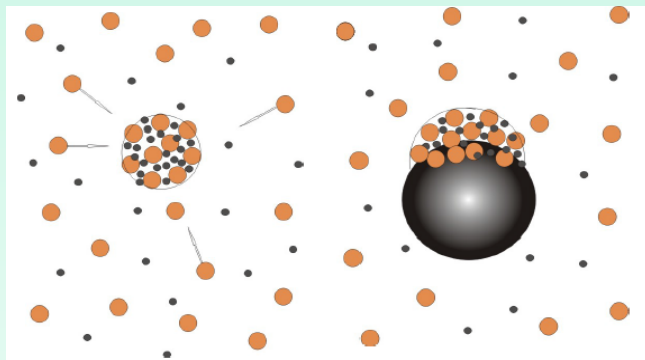


Figure by Hanna Vehkamäki

The substrate lowers the positive free energy term associated to the creation of a spherical surface in the homogeneous case.

Homogeneous Nucleation rate J

number of critical thermodynamically stable cluster (TSC) formed by unit of time and volume

Following the classical approach:

$$J_h = \beta \cdot Z \cdot \left[n_1 \exp \left[\frac{\Delta G^*(p, T)}{kT} \right] \right]$$

growth rate of c.cluster

Zeldovich factor

Describe the prob of stable cluster to re-dissolve (fluctuation around stable state)

Concentration of critical clusters

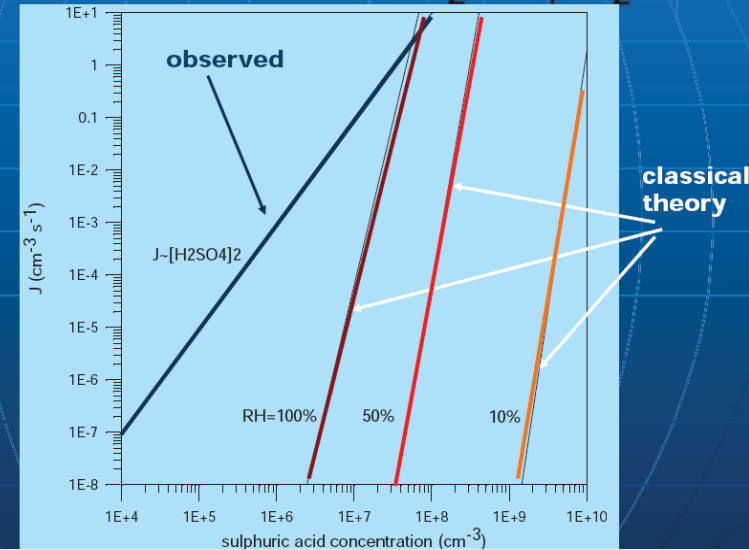
Mechanism usable in atmospheric model (BHN)
nucleation mechanism (use of look up table)

Limitations of the classical theory

-Macroscopic theory for microscopic phenomena (e.g. what is a surface at this scale ?)

-Nucleation is highly local but describe with the macroscopic state of parent phase ... neglect local fluctuations effects ...

Measured and calculated nucleation rates for H₂SO₄-H₂O



Alternatives:

- Kinetic approaches (cf e.g. Hegg et al., 2009)
- Molecular dynamic simulations (probably the most rigorous)
...but not for inclusion in atmospheric models ...

For atmospheric modeling: BHN or use of empirical relationships based on observations *

$$J_2 = A [\text{H}_2\text{SO}_4], \quad (4)$$

$$J_2 = K [\text{H}_2\text{SO}_4]^2, \quad (5)$$

$$J_2 = A_{\text{org}} [\text{NucOrg}], \quad (6)$$

$$J_2 = K_{\text{org}} [\text{NucOrg}]^2, \quad (7)$$

$$J_2 = A_{s1} [\text{H}_2\text{SO}_4] + A_{s2} [\text{NucOrg}], \quad (8)$$

$$J_2 = K_{\text{het}} [\text{H}_2\text{SO}_4] \times [\text{NucOrg}], \quad (9)$$

$$J_2 = K_{\text{SA1}} [\text{H}_2\text{SO}_4]^2 + K_{\text{SA2}} [\text{H}_2\text{SO}_4] \times [\text{NucOrg}], \quad (10)$$

$$J_2 = K_{s1} [\text{H}_2\text{SO}_4]^2 + K_{s2} [\text{H}_2\text{SO}_4] \times [\text{NucOrg}] + K_{s3} [\text{NucOrg}]^2, \quad (11)$$

*Limit of observation ~ 3nm

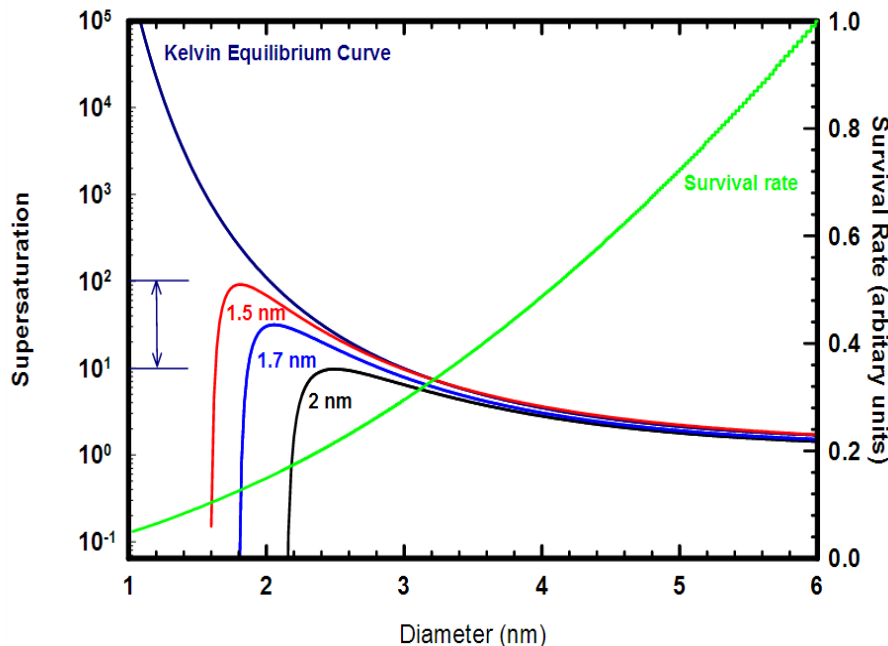
ex: Intercomparison based on wrf-chem : Lupascu et al., ACP, 2015

Activation for growth of the aerosol

Example: Freshly-nucleated thermodynamically stable clusters (TSCs), composed of ammonium bisulfate and water, are activated for condensational growth by a water-soluble organic vapour.

- Activation means that the organic vapor starts to condense irreversibly into these particles allowing growth and further formation of CCN
- However, this does not occur before TSCs have reached a certain size threshold $>$ than the fresh TSC

nano-Köhler (Kulmala et al.)




- Inorganic TSCs 1-3 nm
- activating vapors: soluble
- organic compounds
- 10^6 - 10^8 molec/cm³
- saturation ratio not limited
- S depends on condensation sink, vapors source rate and saturation, fluctuations

fits better the measurements than BHN only

Nucleation rate for a distribution of particles

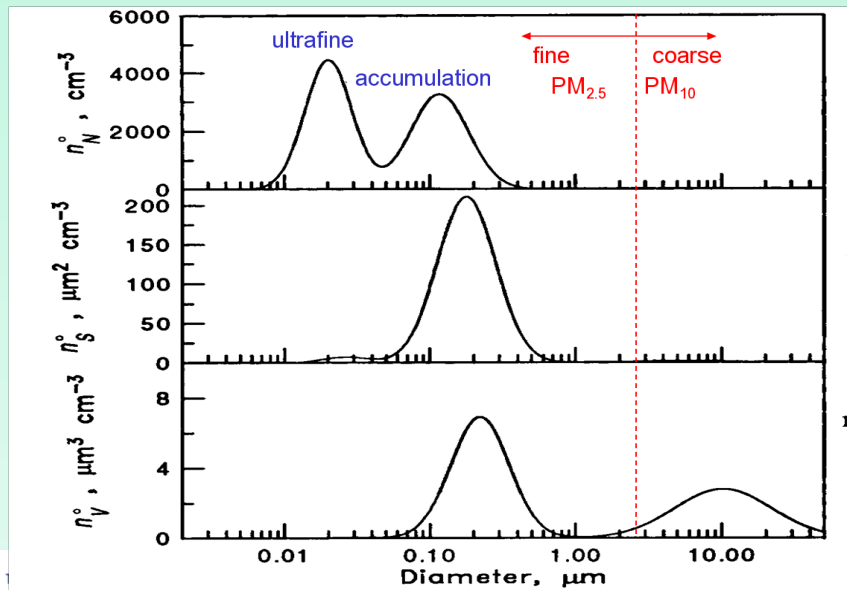
$$\left[\frac{\partial n_i(v, t)}{\partial t} \right]_{nuc} = J_i(v) \delta(v - v_0)$$

source term



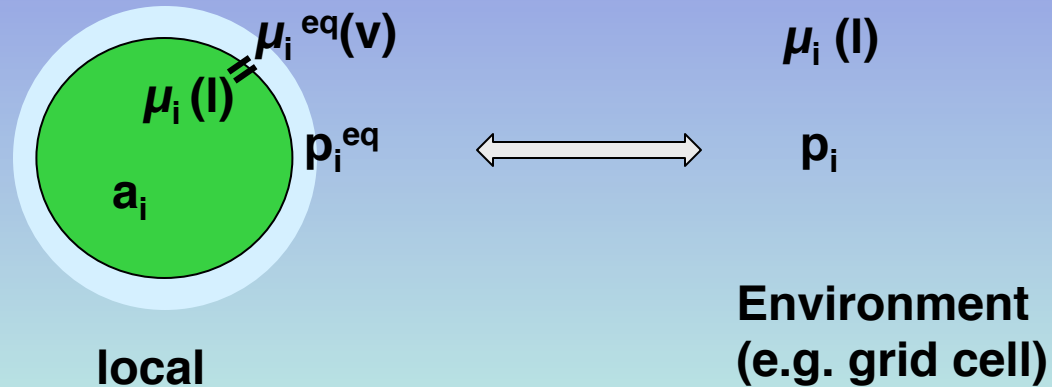
Critical size for growth by condensation (

The “nucleation” mode



Condensation/Evaporation

Condensation rate:
variation of particle size due
to condensation / evap.



$$I(v) = \frac{D_i M_i}{RT} \left[48 \pi^2 \cdot v \right]^{1/3} \cdot f(Kn, \alpha) \cdot (p_i - p_i^{eq}) \quad [\text{m}^3 \cdot \text{s}^{-1}]$$

p_{eq} is calculated with a thermodynamic module solving the chemical composition (activities a_i) of the solution (e.g ISORROPIA). Possibly accounting for the Kelvin effect (for smaller particles) on p_{eq} .

f accounts for the transition regime between kinetic and continuum regimes based on *Knudsen number* (ratio of mean molecular free path to particle radius D_p) and the *accommodation coefficient* (probability of the molecule to “stick” to the condensed phase).

Condensation rate for a distribution of particles

$$\left[\frac{\partial n_i(v, t)}{\partial t} \right]_{\text{cond/evap}} = - \frac{\partial}{\partial v} [I_i(v) n_i(v, t)]$$

Diffusion growth equation can be solved for multiple component with appropriate numerical methods (cf Jacobson, Fundamentals of Atmospheric Modelling, APC analytical predictor of condensation).

Equilibrium time scale : characteristic time for p_i to reach p_{eq} vary from second to days, very sensitive to particle radius. Large aerosol are slower to reach equilibrium.

Model simplifications:

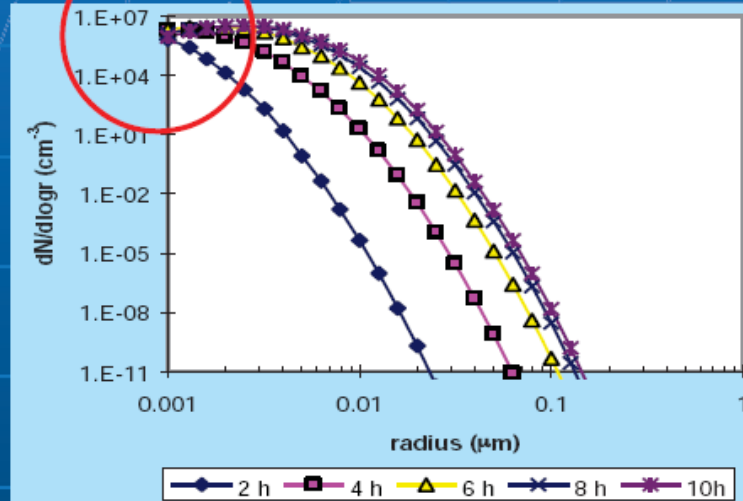
If model time step $\Delta t \gg \tau_{eq}$ for small particles (bin or modes) consider that P_{eq} is reached instantaneously and do not solve the kinetic equation. This approximation cannot be used to treat cond/evap from larger particles.

In bulk models:

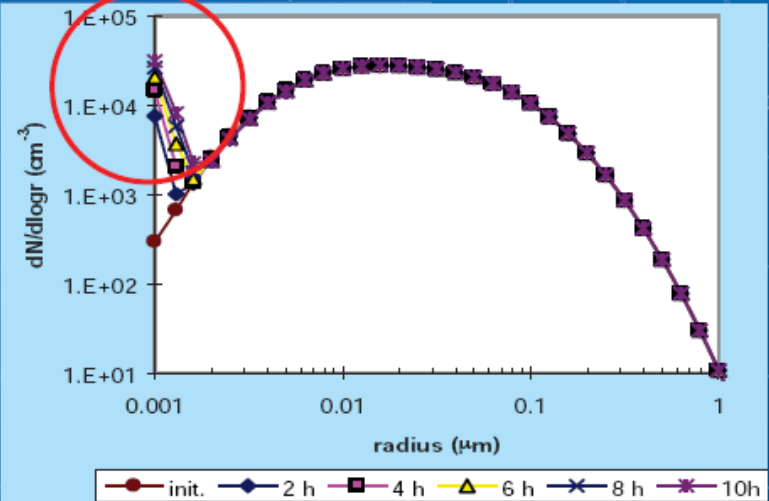
Use only thermodynamical equilibrium to treat gas / particle partition.

Use some empirical scavenging coefficients

Competition between nucleation and condensation



Nucleation without pre-existing particles



Nucleation in presence of pre-existing particles

**Nucleation is less efficient!!!
Condensation subtracts gas**

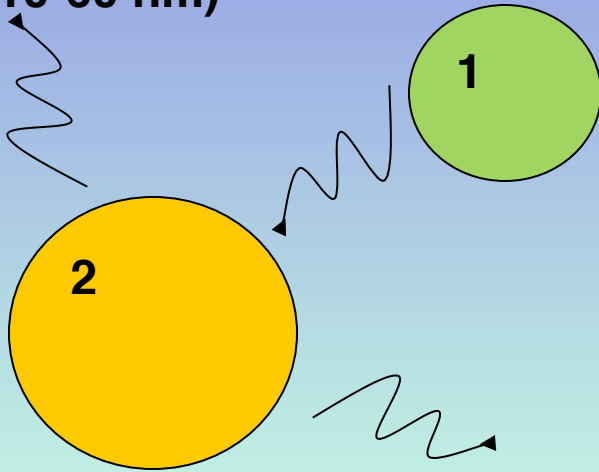
Coagulation

It is the process by which particles collide among them due to their relative motion and stick to form a larger particle :

- **Thermal coagulation : driven by Brownian motion**
- **Kinematic coagulation : driven by external actions: gravity, electric forces, aerodynamical effects ...**

Brownian coagulation coefficients

Aerosol mean free path
(10-60 nm)



Aerosol Brownian diffusivities (Stokes-Einstein)
is a determining factor

$$K_{1,2} = \frac{2kT}{3\mu} \frac{(v_1^{1/3} + v_2^{1/3})}{(v_1 v_2)^{1/3}} \cdot \beta$$

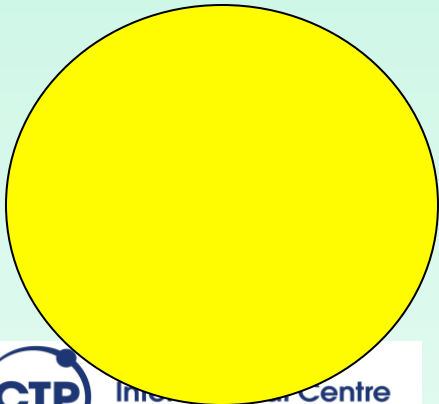
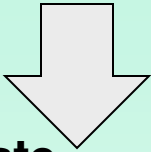
μ : air viscosity

β : Fuchs correction factor : accounts for collision efficiency and other effects, depends on Kn (diffusing part. free path / absorbing particle radius).

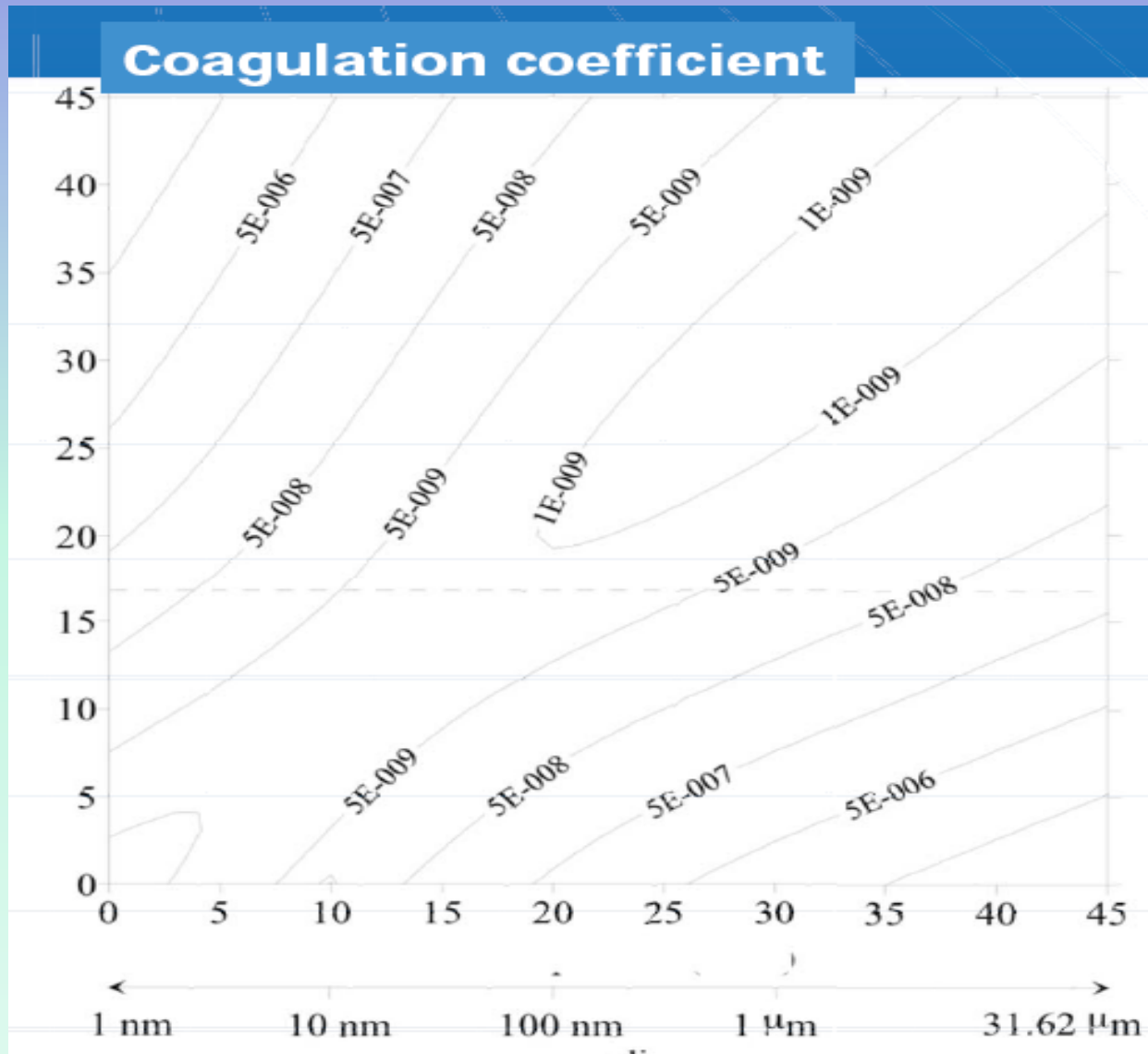
Kn \rightarrow 1, β (Kn) : free molecular regime ($D_p < 0.1$ micron)

Kn \rightarrow 0, $\beta \rightarrow$ 1: continuum regime ($D_p \sim > 1$ micron)

$J = K_{1,2} N_1 N_2$
Formation rate



Particle 2



Particle 1

Kinematic coagulation processes vs. Brownian coagulation:

Effect of laminar shear and settling velocity

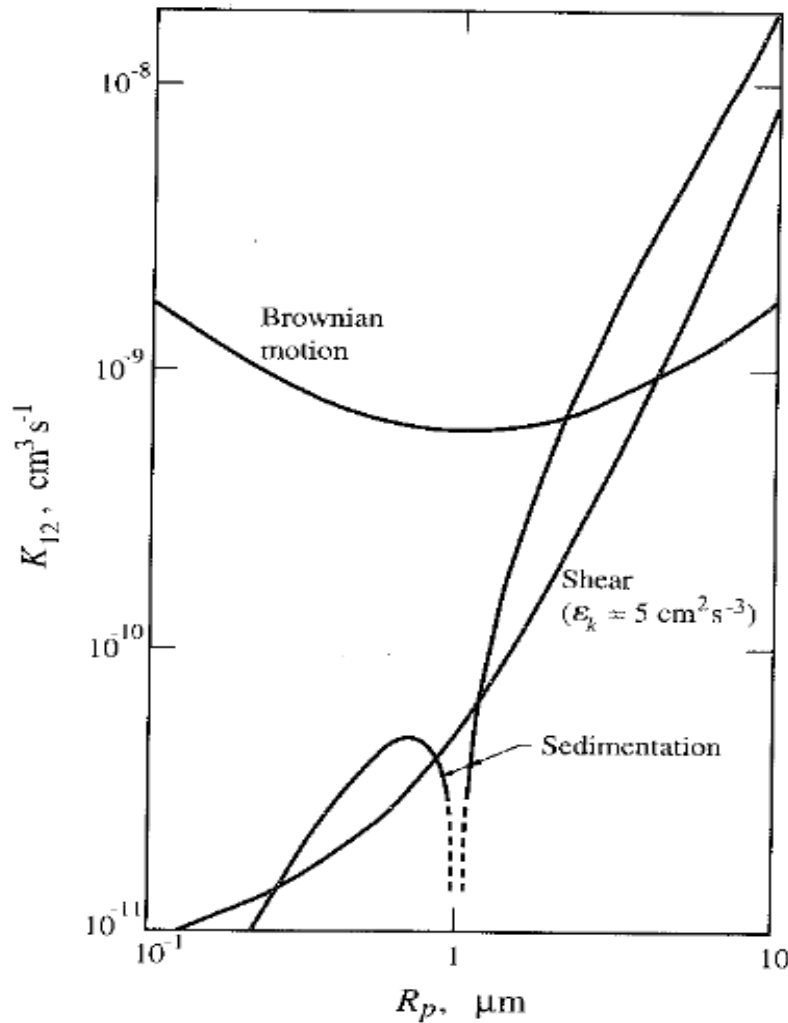


FIGURE 12.7 Comparison between coagulation mechanisms for a particle of $1 \mu\text{m}$ radius as a function of the particle radius of the second interacting particle.

From Seinfeld and
Pandis

Effect of external force fields on coagulation coefficients

Wan der Walls forces (momentary dipoles forming)

Coulomb forces if charged particles

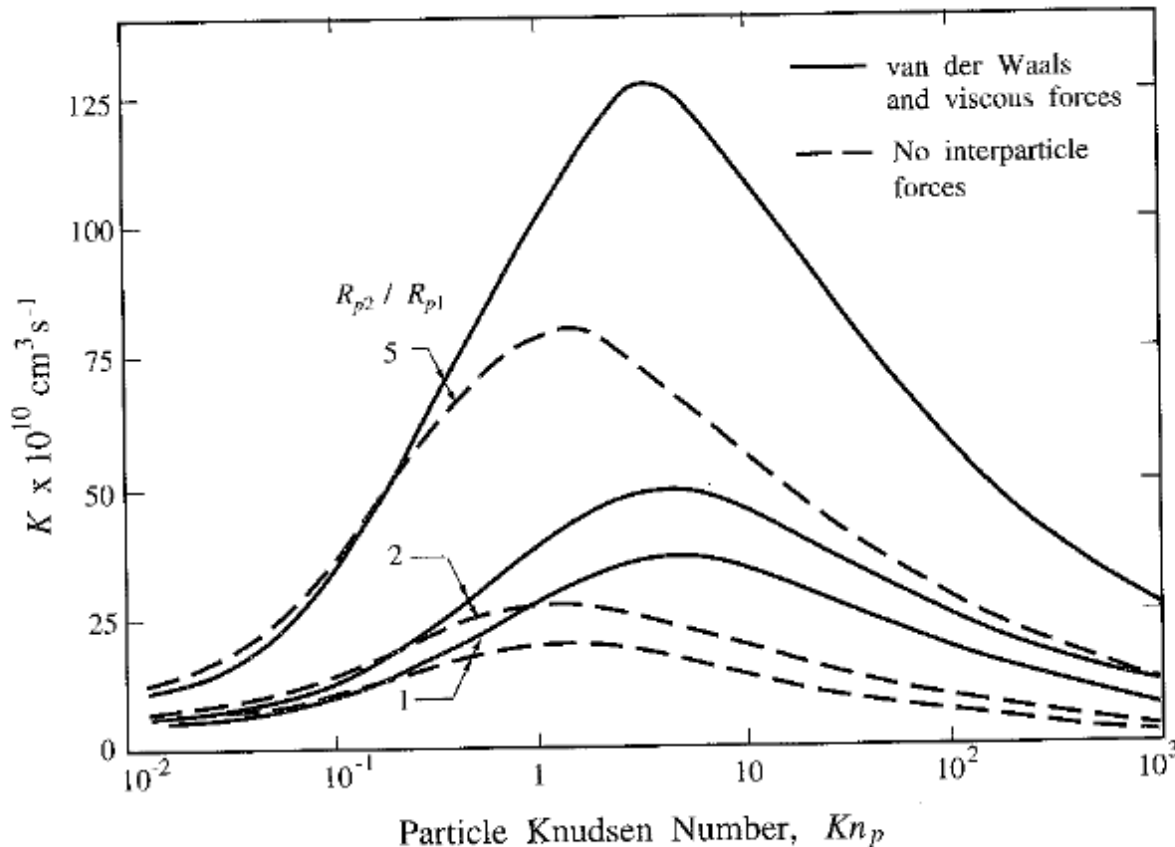


FIGURE 12.10 Coagulation coefficient at 300 K, $\rho_p = 1 \text{ g cm}^{-3}$, $A/kT = 20$ as a function of Knudsen number (Kn_p) for particle radii ratios of 1, 2, and 5 both in the presence and in the absence of interparticle forces.

Coagulation rate for a distribution of particles

$$\left[\frac{\partial n_i(v, t)}{\partial t} \right]_{coag} = \frac{1}{2} \int_0^v K(v-q, q) n(v-q, t) n(q, t) dq - n(v, t) \int_0^\infty K(q, v) n(q, t) dq$$



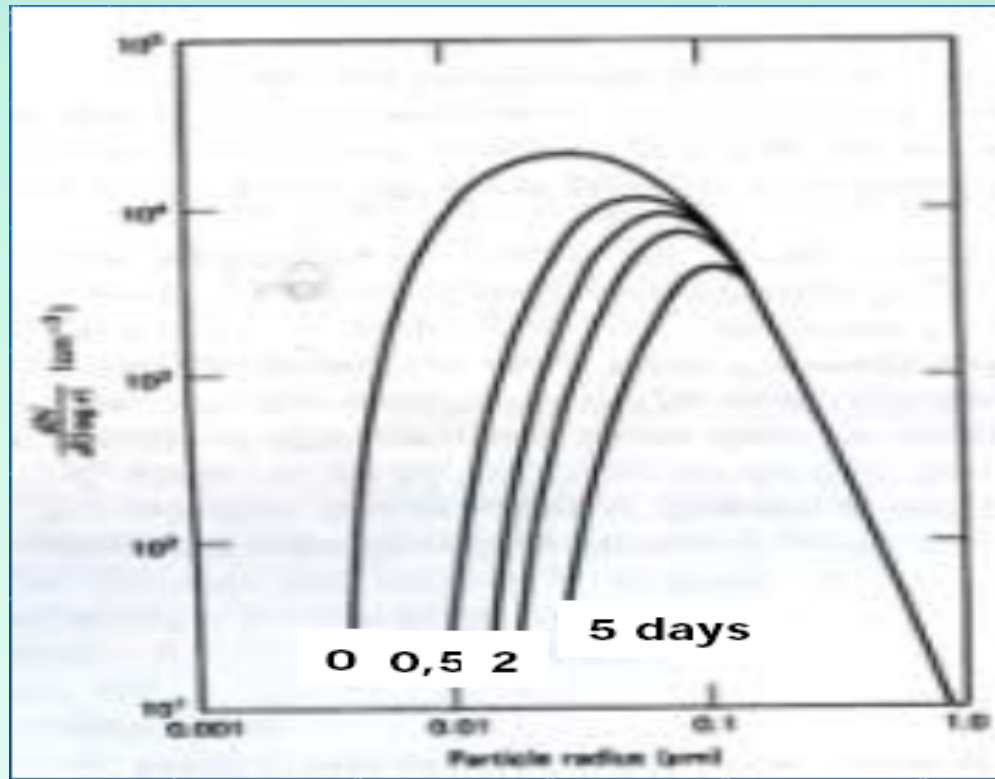
Source term for the particle
number of volume range $v, v+dv$



Sink term for the particle
number of volume range $v, v+dv$

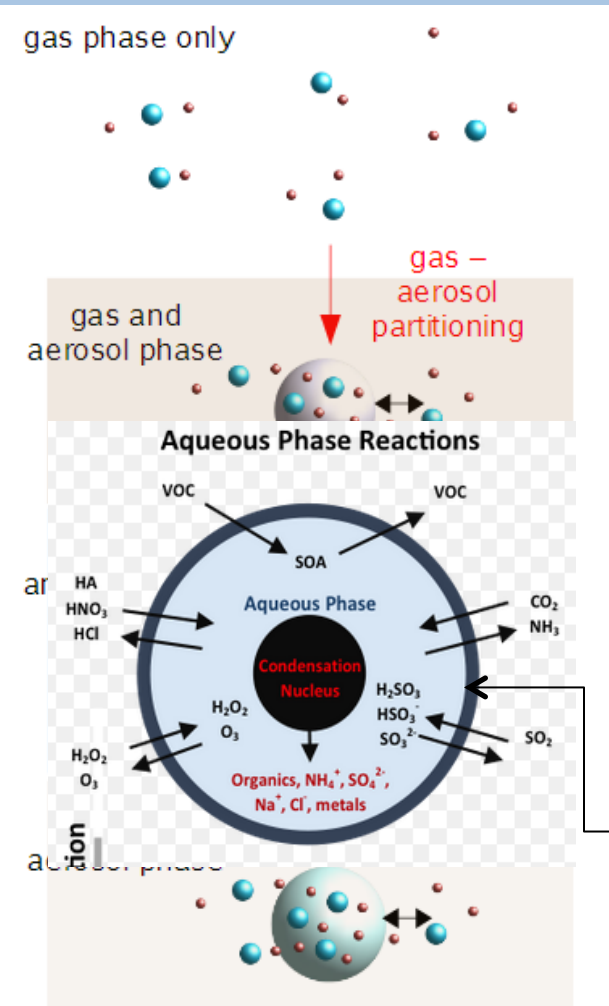
Overall the mass is conserved

Typical evolution of size
distribution due to coagulation



Aerosol - Cloud processing

Non precipitating cloud water



Picture from H. Tost

~ On average 10% of a forming cloud is precipitated

The recycling usually increase the size mass of the initial aerosol. Via in cloud conversion of gas phase material.

On aerosol can undergo several cycles

Very important pathway for sulfate formation SO₂ oxidation by H₂O₂ and O₃ in aqueous phase.

Modelling is complex (clouds!). Needs aqueous chemistry solver to estimate the gain of mass.

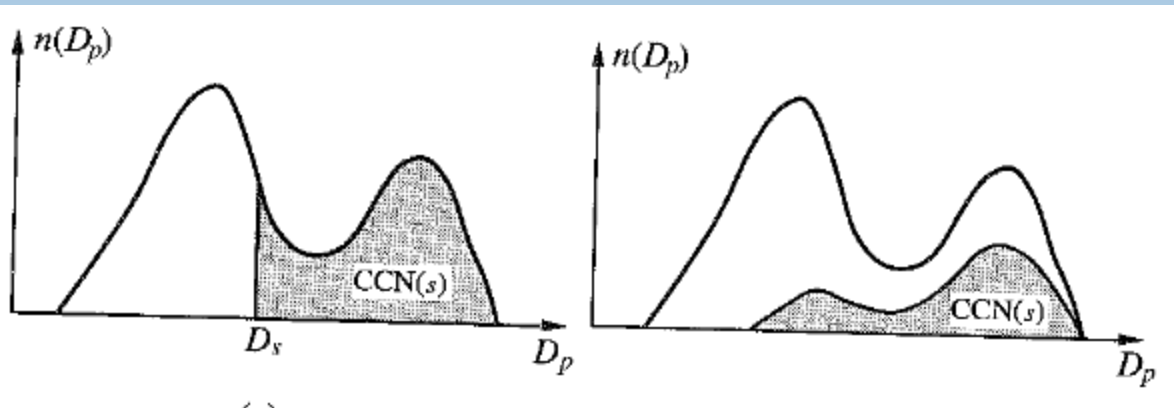
Hoose et al., ACP, 2008; Gong et al., Atmosphere, 2011 examples of param.

+ capture of interstitial particles
+ In cloud coalescence

Slow processes

Aerosol wet scavenging : rainout

Rainout : results from the conversion of nucleated cloud droplet to rain droplets falling onto the surface (if no re-evaporation).



Bulk schemes use the cloud to rain water auto-conversion rate.

More evolved schemes track the “aerosol in cloud” as a prognostic species (Hoose et al., ACP, 2008) and represent the effect on cloud processing (including removal) on aerosol size distribution.

Aerosol wet scavenging : washout

Collection of particles (dp) by falling meteors

Scavenging coefficient $\Lambda(dp) = \int_0^{\infty} \frac{\pi}{4} D^2 U_t(D) E(D, dp) N(D) dD$

$N(D)$: size distribution of falling meteors

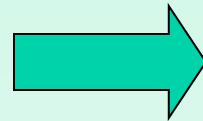
$U_t(D)$: terminal velocity of falling meteor

$N(D)$: number concentration of fall

E : Collection efficiency
(see Slinn, 1983 and Seinfeld and Pandis)
=1 if all the particles in the volume swept out by the droplet are collected.

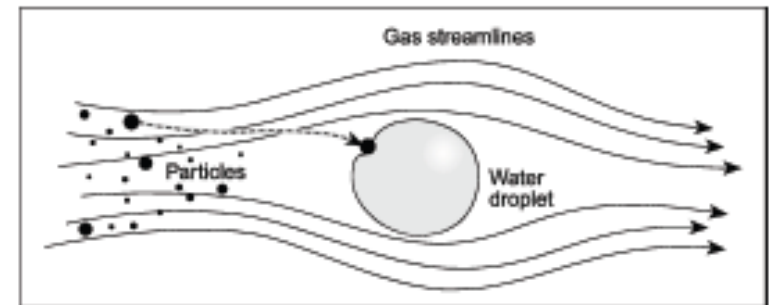
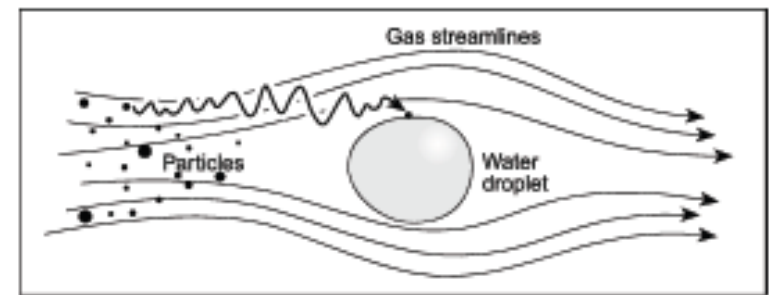
... a complex problem...

Qualitatively

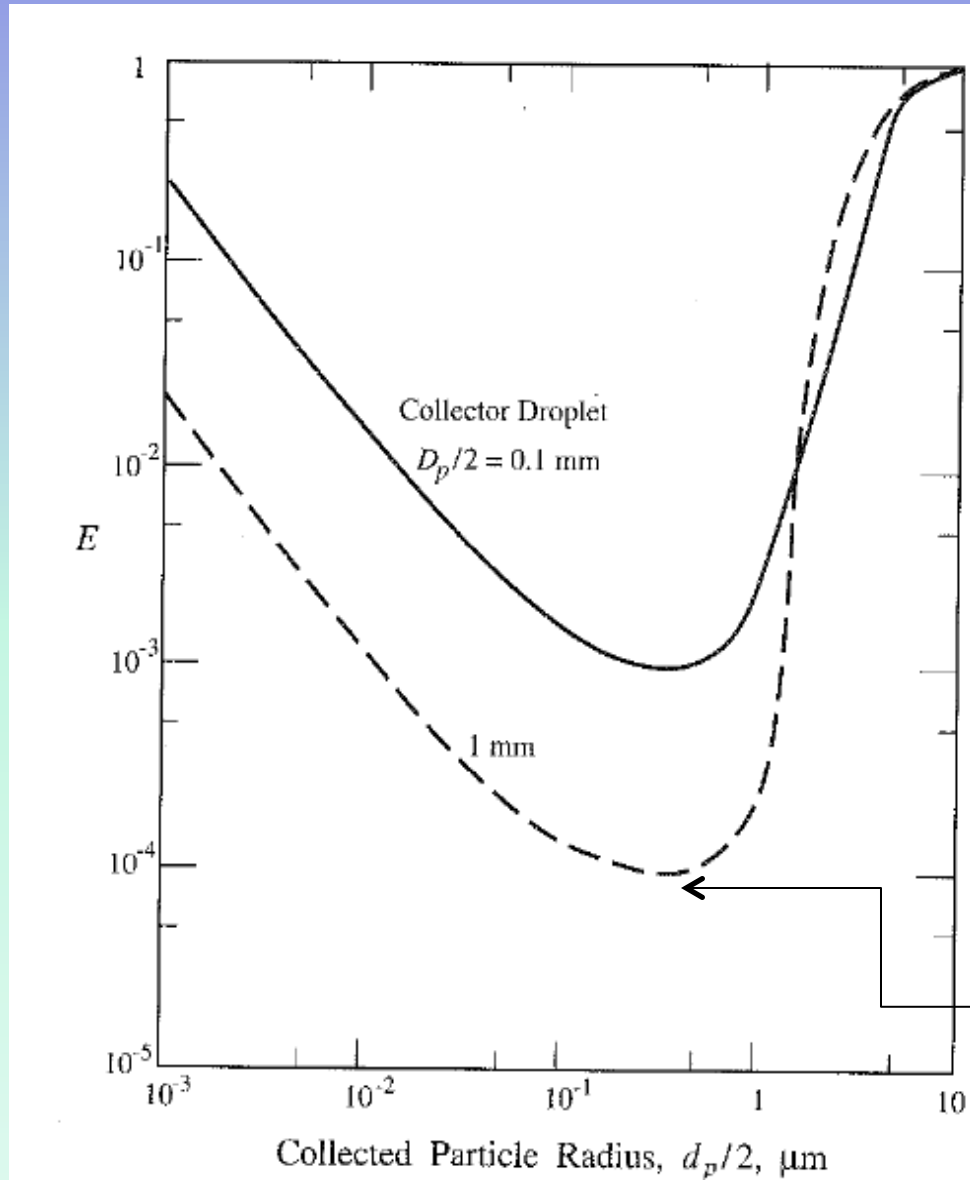


-Brownian diffusion

-Inertial impaction



Collection efficiency as a function of particle and hydrometeor size



"Greenfield gap"

Rainout on a population of aerosol

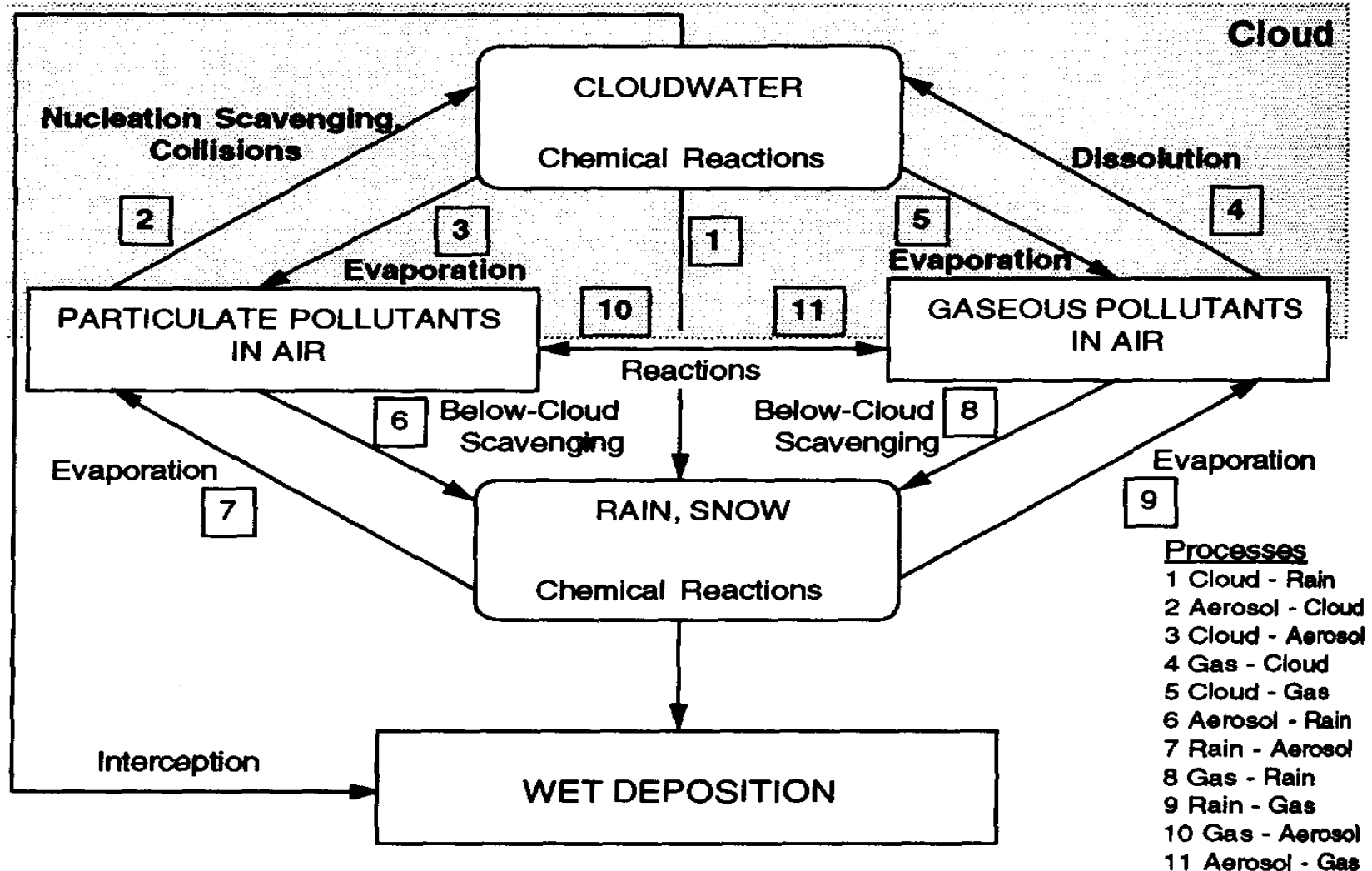
$$\left[\frac{\partial n_i(v, t)}{\partial t} \right]_{rainout} = -\Lambda(v, t) \cdot n_i(v, t)$$

The double size integration (particles/hydrometeor) requires a size resolving rain scheme.

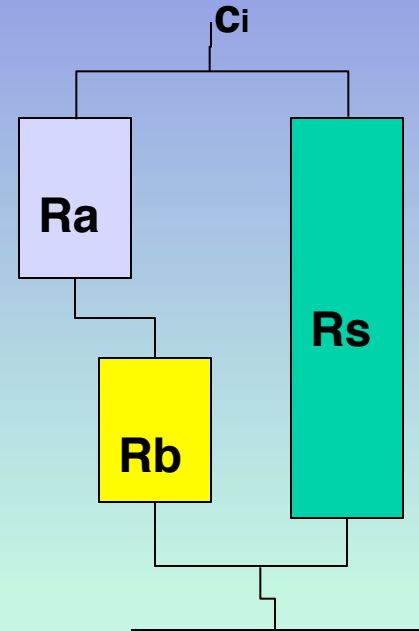
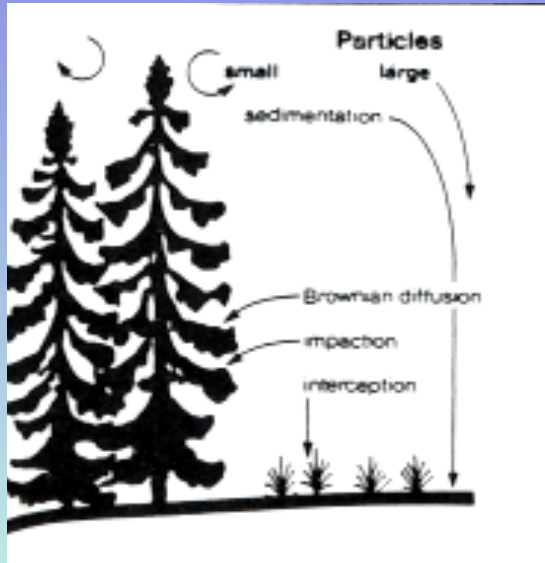
Often a typical size distribution for rain is assumed (e.g. Marshall-Palmer)

When working with a bulk scheme (aerosol mass), need to assume a typical size distribution for aerosol too

Overview of cloud porcessing (Seinfeld and Pandis)



Dry deposition velocity



Assuming perfect interception

$$Vd_i = \frac{1}{Ra + Rb_i + RaRb_iVs_i} + Vs_i$$

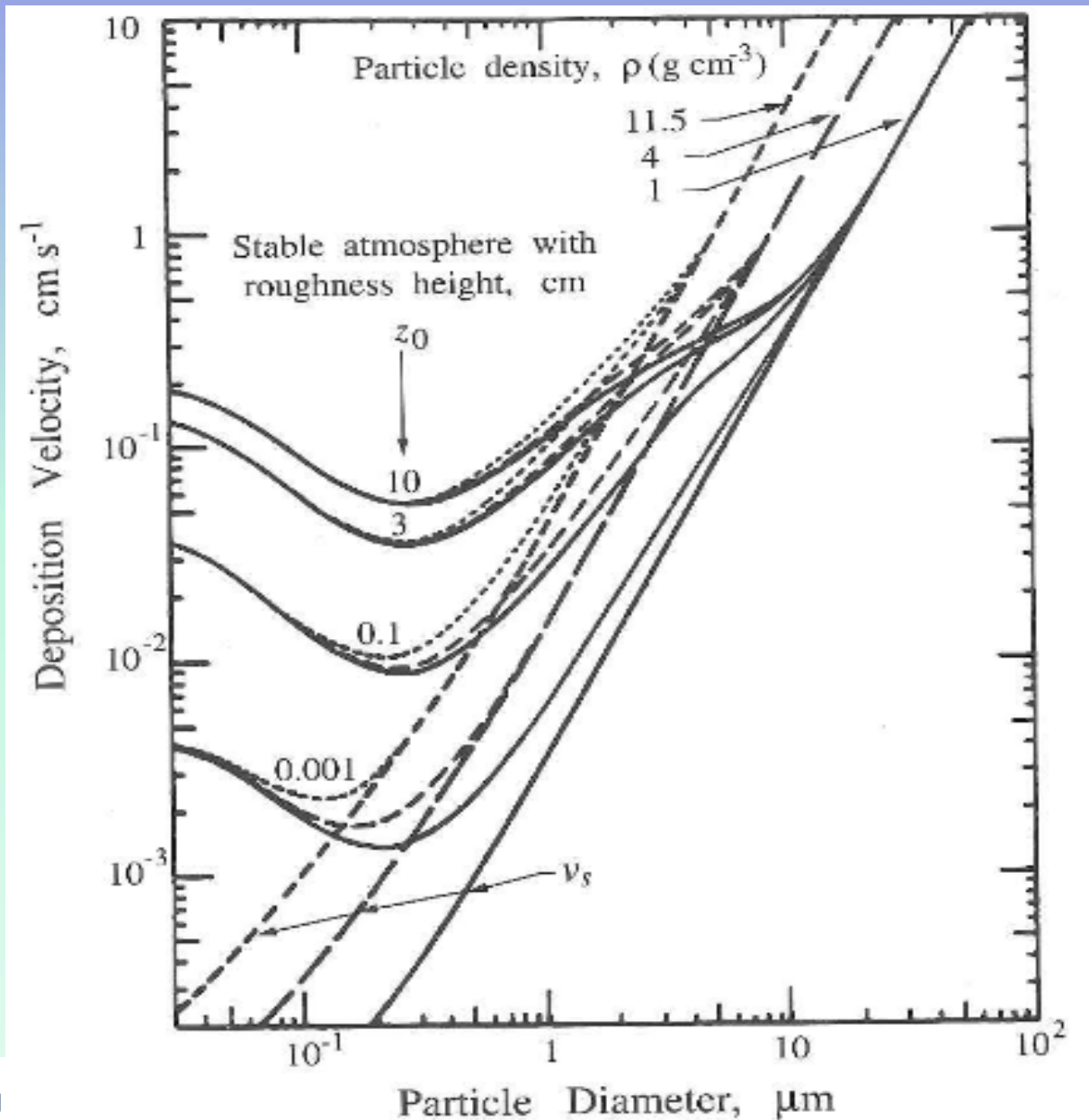
Controlled by
turbulence and
surface roughness

Controlled by particle size and
density, air characteristics

Controlled by particle size and
density (brownian/inertia), air
characteristics in the laminar


Alternative param.
Zhang et al., ACP 2014

Deposition velocity, size and roughness dependence



Effect on the size distribution

$$\left[\frac{\partial n_i(v, t)}{\partial t} \right]_{sed / drydep} = - \frac{V d_i(v, t)}{\Delta z} \cdot n_i(v, t) \quad \Rightarrow \quad \text{Accumulation mode}$$



 Surface layer height

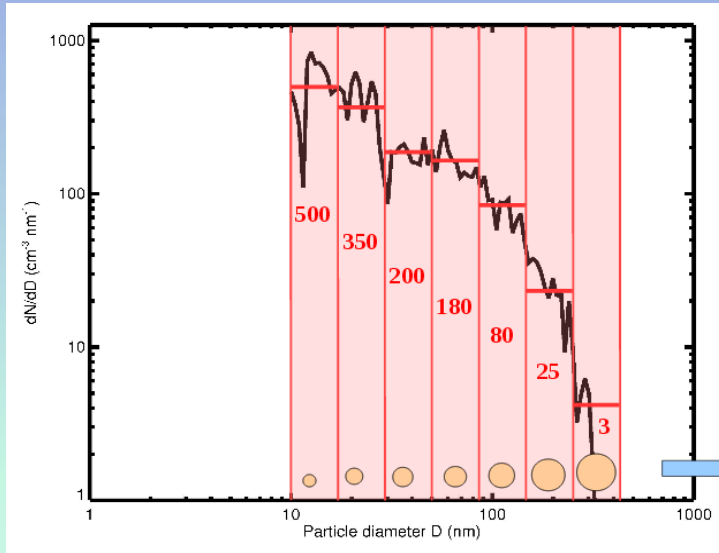
➡ In higher layer consider calculating the sedimentation velocity tendency as the divergence of sedimentation flux.

Table 20.2 Time for a particle (or a gas for the smallest size) to fall 1 km in the atmosphere by sedimentation

Diameter (μm)	Time to fall 1 km	Diameter (μm)	Time to fall 1 km
0.0005	9630 y	4	23 d
0.02	230 y	5	14.5 d
0.1	36 y	10	3.6 d
0.5	3.2 y	20	23 h
1	328 d	100	1.1 h
2	89 d	1000	4 m
3	41 d	5000	1.8 m

Two strategies for representing size evolution

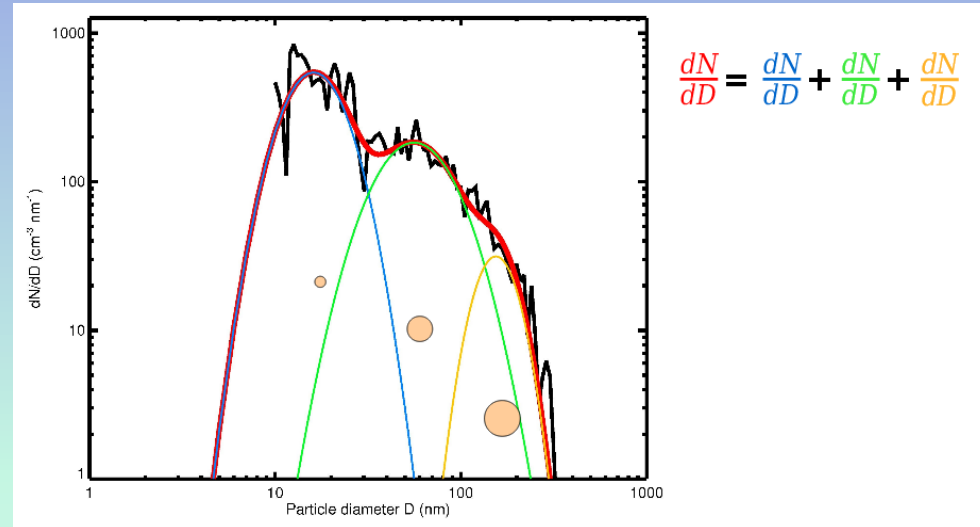
Sectional approach (size bins)



Discretize the distribution with fixed or dynamic size bins.

Can become expensive if we want to keep the accuracy
number of tracer =
nbin*nspecies

Modal approach / moment method



Assume a size distribution e.g. log-normal

$$n_N(D_p) = \frac{dN}{dD_p} = \frac{N}{(2\pi)^{1/2} D_p \ln \sigma_g} \exp \left(-\frac{(\ln D_p - \ln \bar{D}_{pg})^2}{2 \ln^2 \sigma_g} \right)$$

$$n_N(r) = \sum_{i=0}^{\infty} \alpha_i M_i$$

Express the terms of the aerosol dynamic equation in terms of the moments of the modes (generally 2 or 3 moments / number, median diameter, variance)

Major types of Aerosols



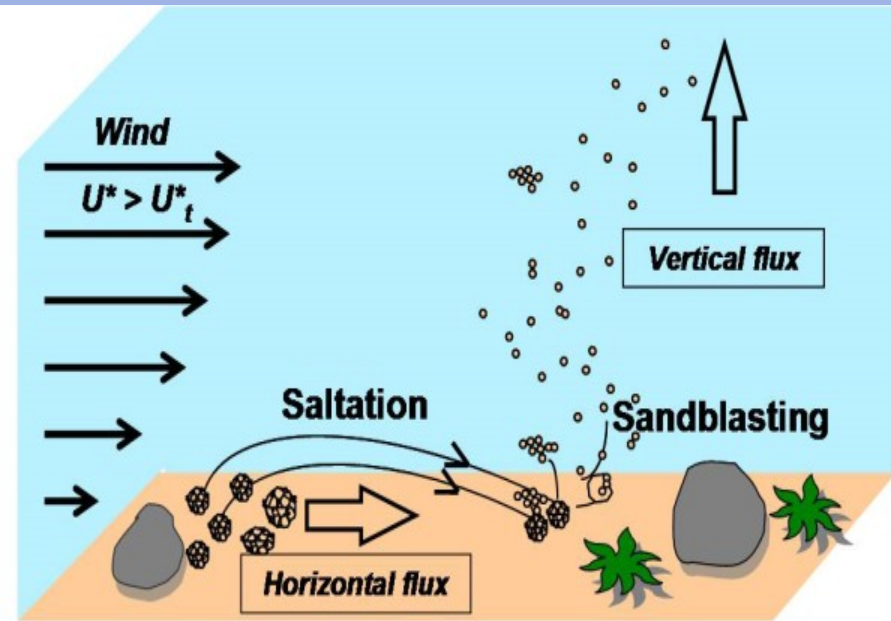
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Natural vs. anthropogenic

Primary vs. secondary

Sources characterizations

Dust emissions processes



Soil texture
Soil humidity
Surface roughness
Surface winds



Threshold friction velocity
Efficiency of sandblasting

$$G = E * \frac{\rho_a}{g} * U^{*3} * \int_{D_p} \left(1 + \frac{U_t^*(D_p, Z_0, z_0)}{U^*} \right) \left(1 - \frac{U_t^*(D_p, Z_0, z_0)^2}{U^{*2}} \right) dS_{rel}(D_p) dD_p$$

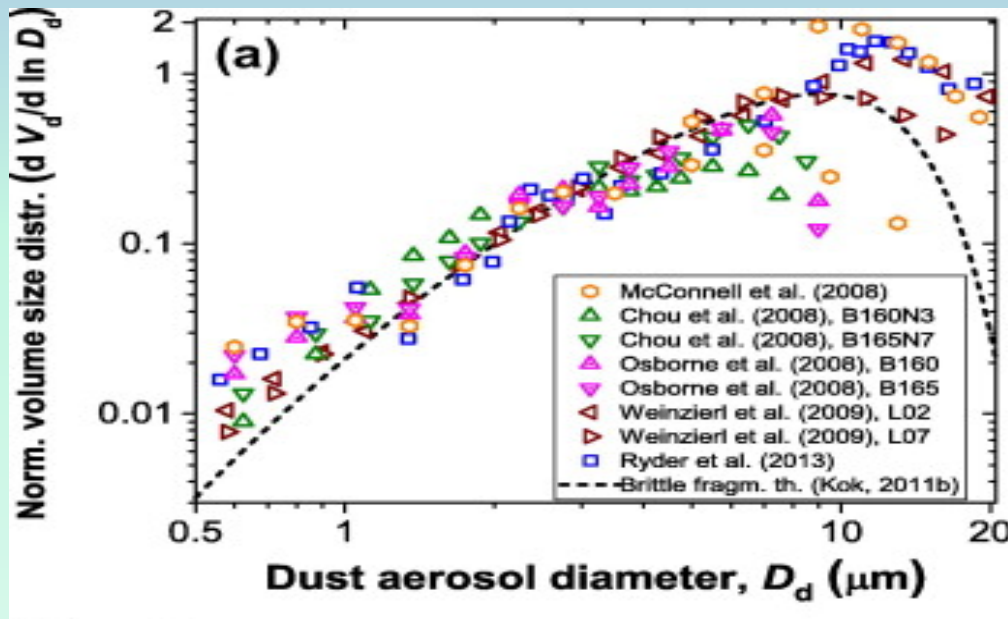
$$\alpha = \frac{F}{G} = c_{alpha} \cdot 10^{0.134 \cdot \%clay - 6}$$

Marticorena and Bergametti,
1995

D_p soil
aggregate
diameter

Dust emission size distribution

- Approaches based on physical model of sand-blasting : problem = parameters, poorly constrained for regional and global applications...
- A recent study suggests that the physics of the scale-invariant fragmentation of brittle material is applicable to dust emission. The resulting emission distribution is independent of u^* .



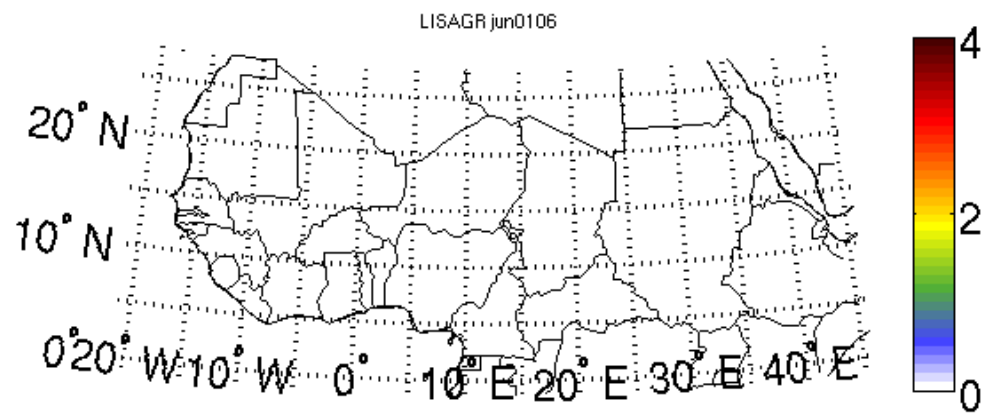
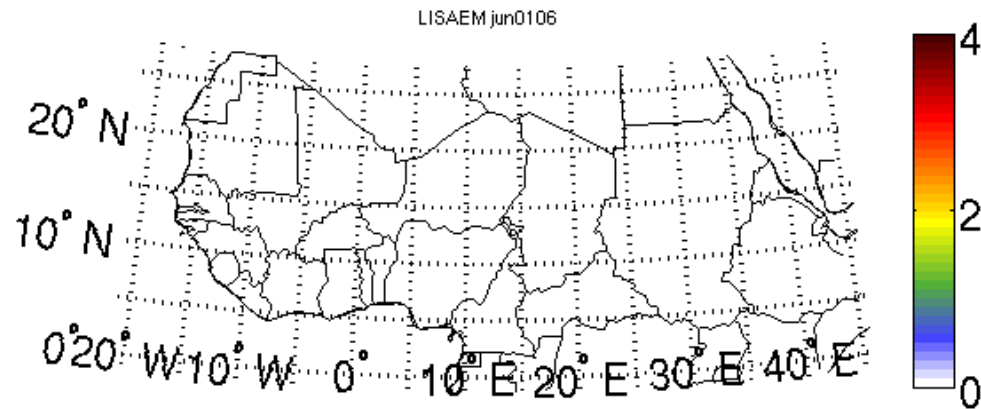
Kok et al., 2011;
Mahowald et al., 2014

$$\frac{dV_d}{d \ln D_d} = \frac{D_d}{c_v} \left[1 + \operatorname{erf} \left(\frac{\ln(D_d/\bar{D}_s)}{\sqrt{2} \ln \sigma_s} \right) \right] \exp \left[- \left(\frac{D_d}{\lambda} \right)^3 \right],$$

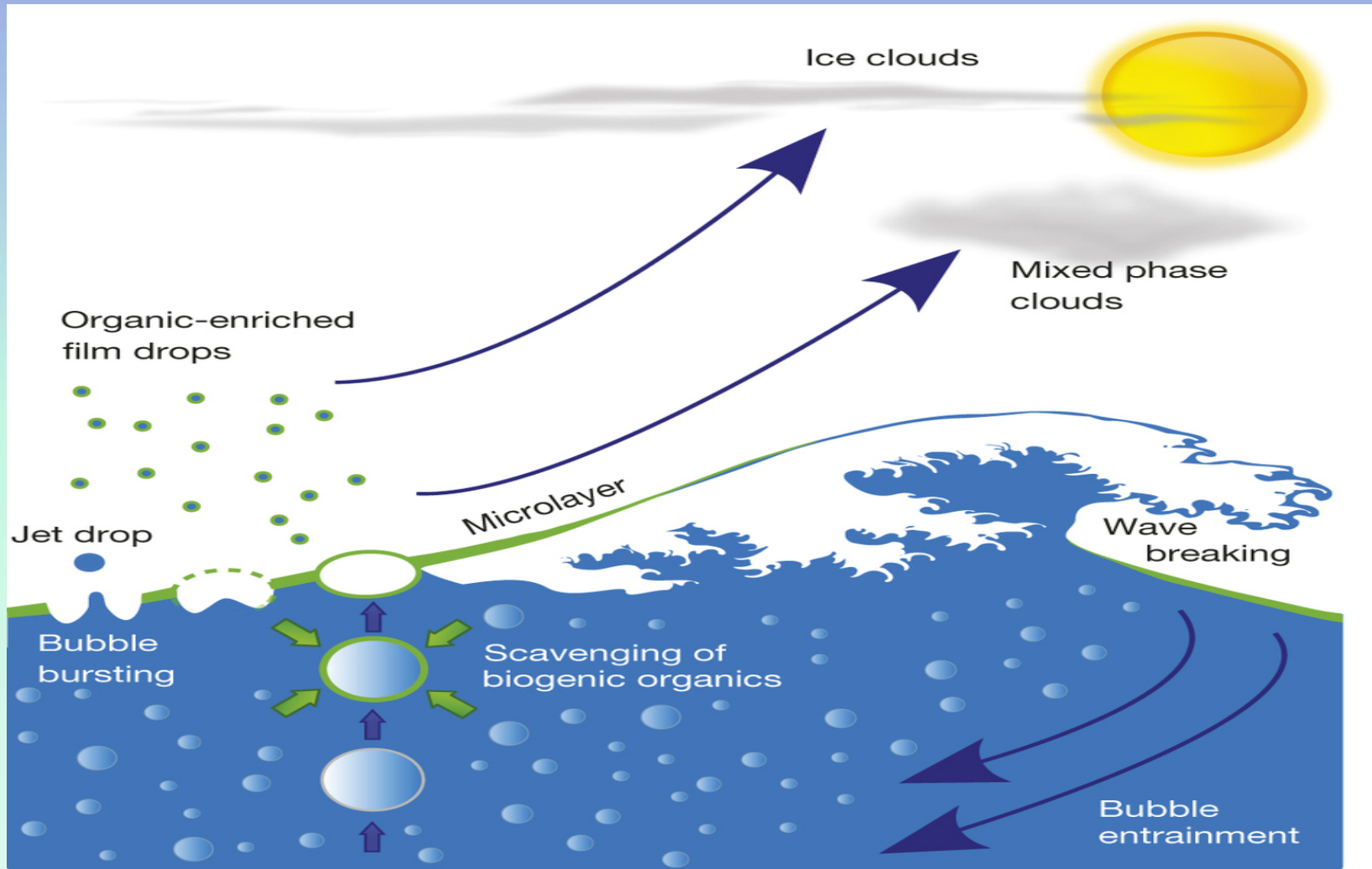
$c_v = 12.62 \mu\text{m}$ is a normalization constant,
 $\sigma_s \approx 3.0$ and $\bar{D}_s \approx 3.4 \mu\text{m}$: log-norm
param of a typical soil particle distrib.

λ : propagation distance **for cracks**,
 $\lambda = 12 \pm 1 \mu\text{m}$

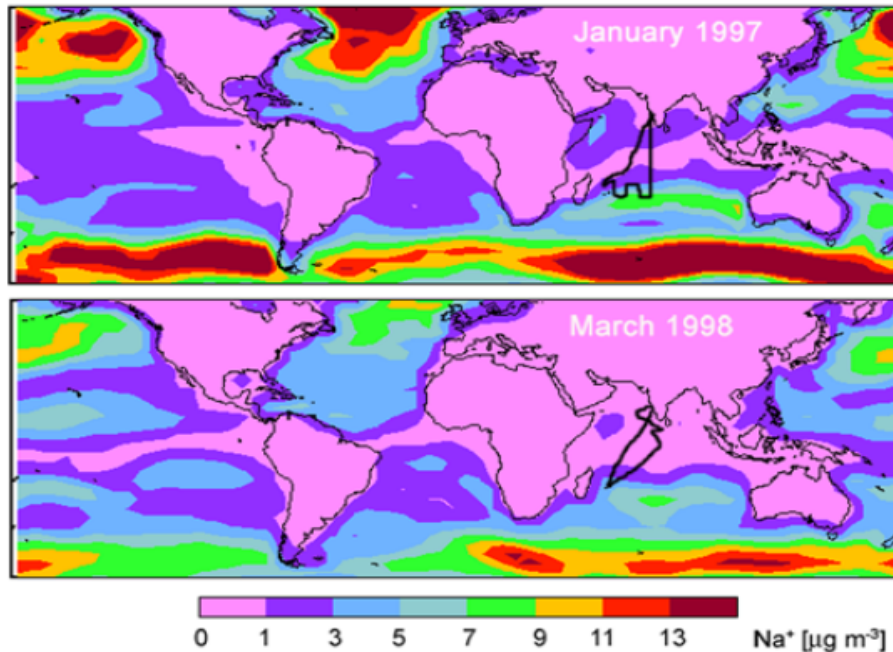
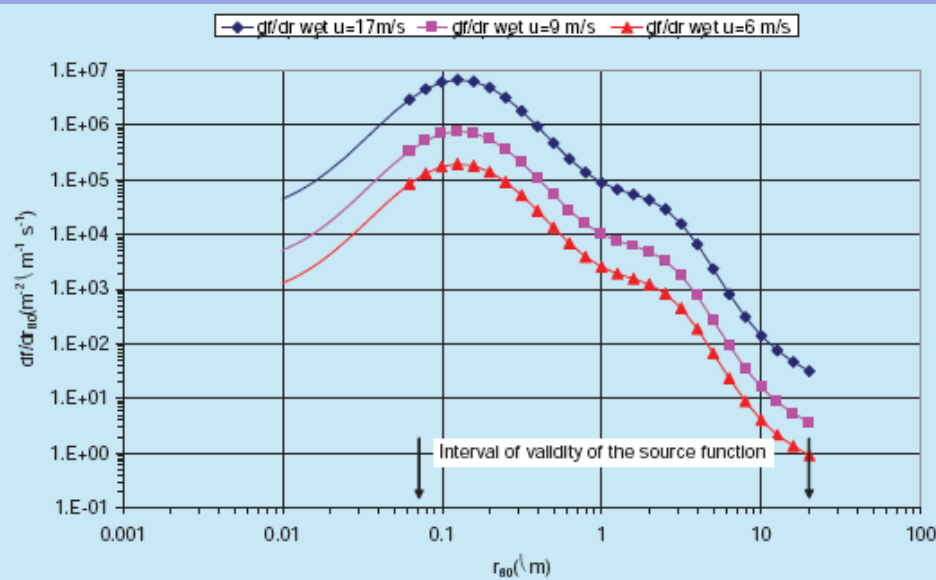
Sensitivity of dust emission to the driving model



Sea Salt and seas sprays aerosol



Sea salt emissions vs wind speed



[Alexander et al. 2005]

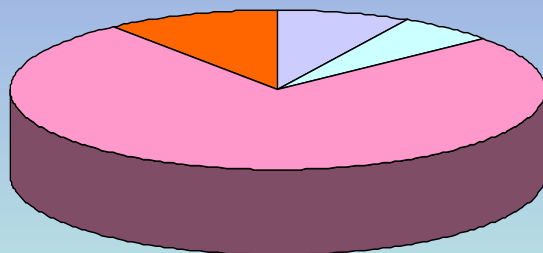
Carbonaceous aerosol

ORGANIC CARBON (OC)

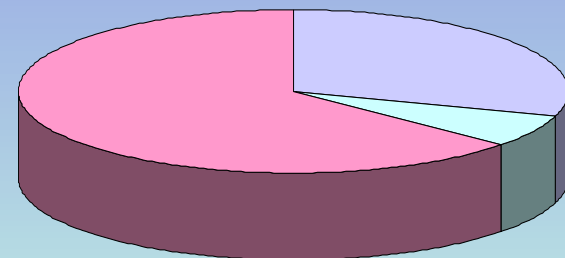
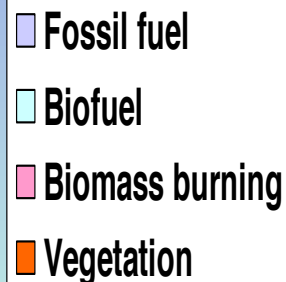
ELEMENTAL CARBON (BC)

Black carbon

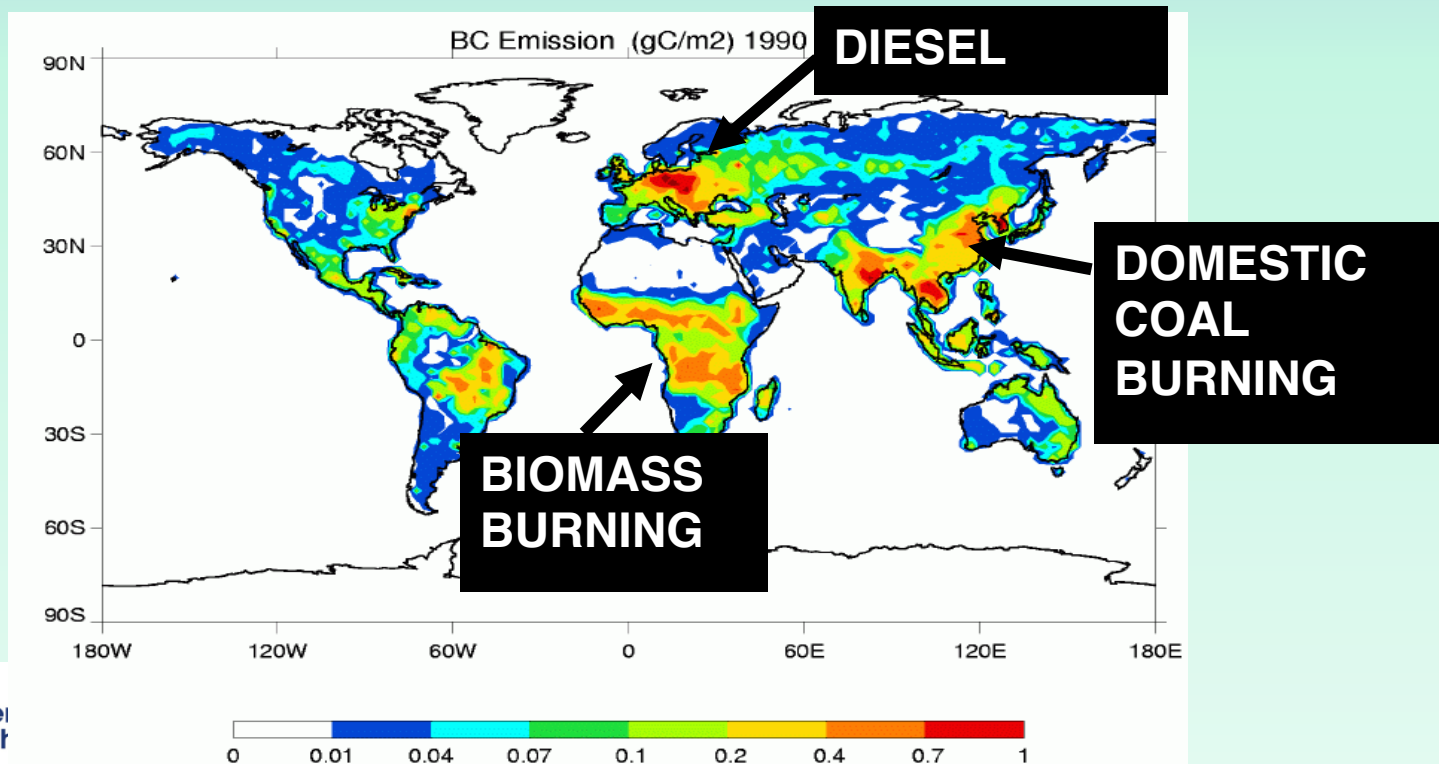
GLOBAL



130 Tg yr⁻¹



22 Tg yr⁻¹



Biomass burning emissions

$$Q(X) = M \times EF(X)$$

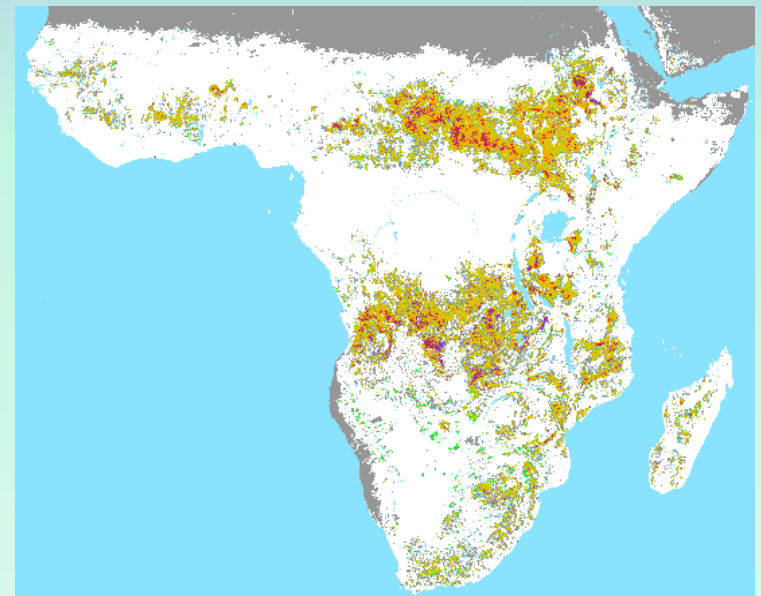
- M is the biomass burned
- $EF(X)$ is the chemical species emission factor X in $gX/kgdm$



M ? for savannah and forest fires

$$M = A \times B \times \alpha \times \beta$$

A is the burned area
 B biomass density
 α , biomass surface fraction
and β , combustion efficiency.



L3JRC

Biomass burning in Earth system models

Anthropogenic emissions

Emission of a compound i ...

$$E_i = E_{traffic} + E_{power} + E_{petro} + E_{ships} + E_{planes} + E_{...}$$

$$E_{i,s} = EF_{i,s} \times N_s \times U_s$$

EF : Emission factors represents the emission potential of a given emitter. The unit vary according to the process e.g : g.km⁻¹ for car traffic sources; g.l⁻¹ for solvent ; g.kg⁻¹ for biogenic emission

N : represent an “quantity of activity” on a given area (e.g a grid cell) - it scales up emission factor to give actual surface fluxes- , e.g. number of km made , quantity of biomass etc ..

U : a fonction modulating the emission typically in fonction of time (U(t)).
Account for diurnal and seasonal variations of emissions, change of technology

Emission size distributions

Typical size distributions for different sector

e.g. Denier van der Gon et al., 2009

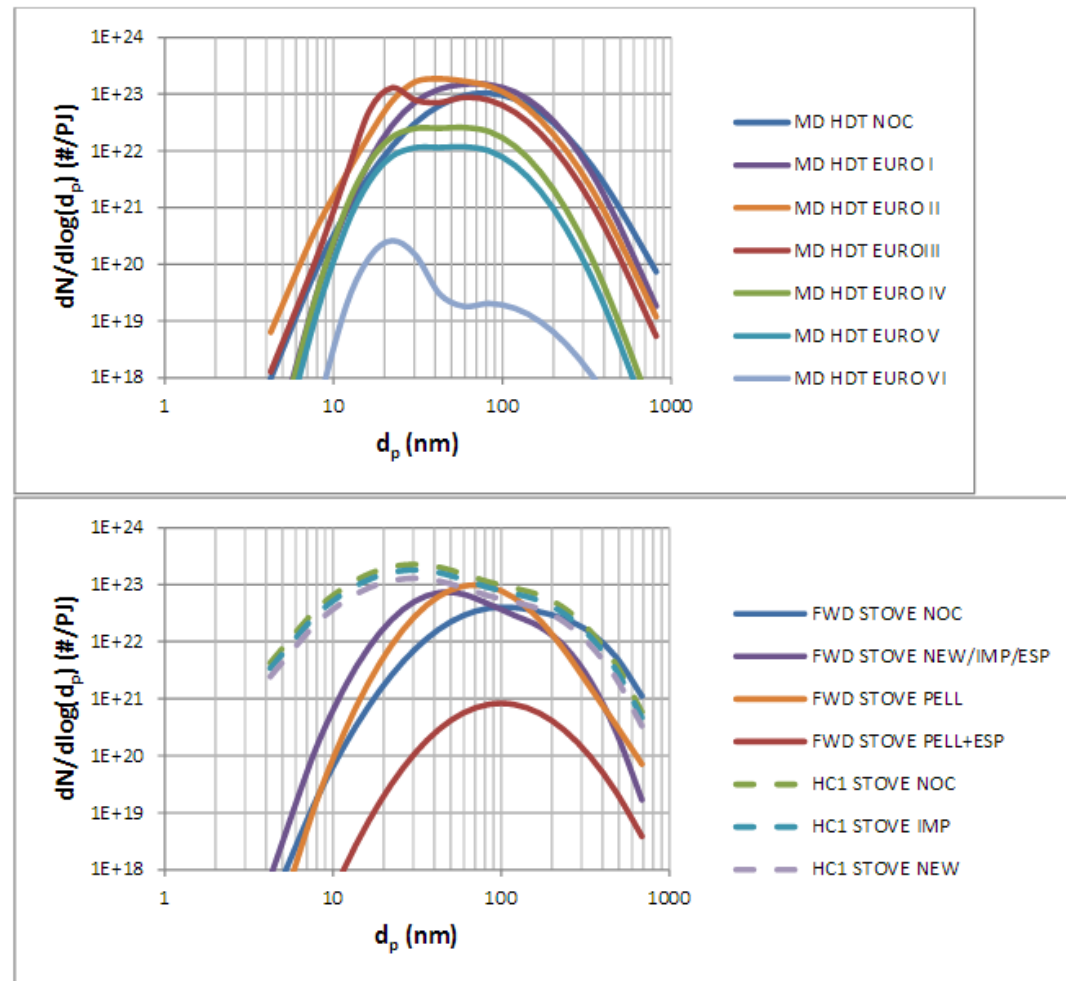
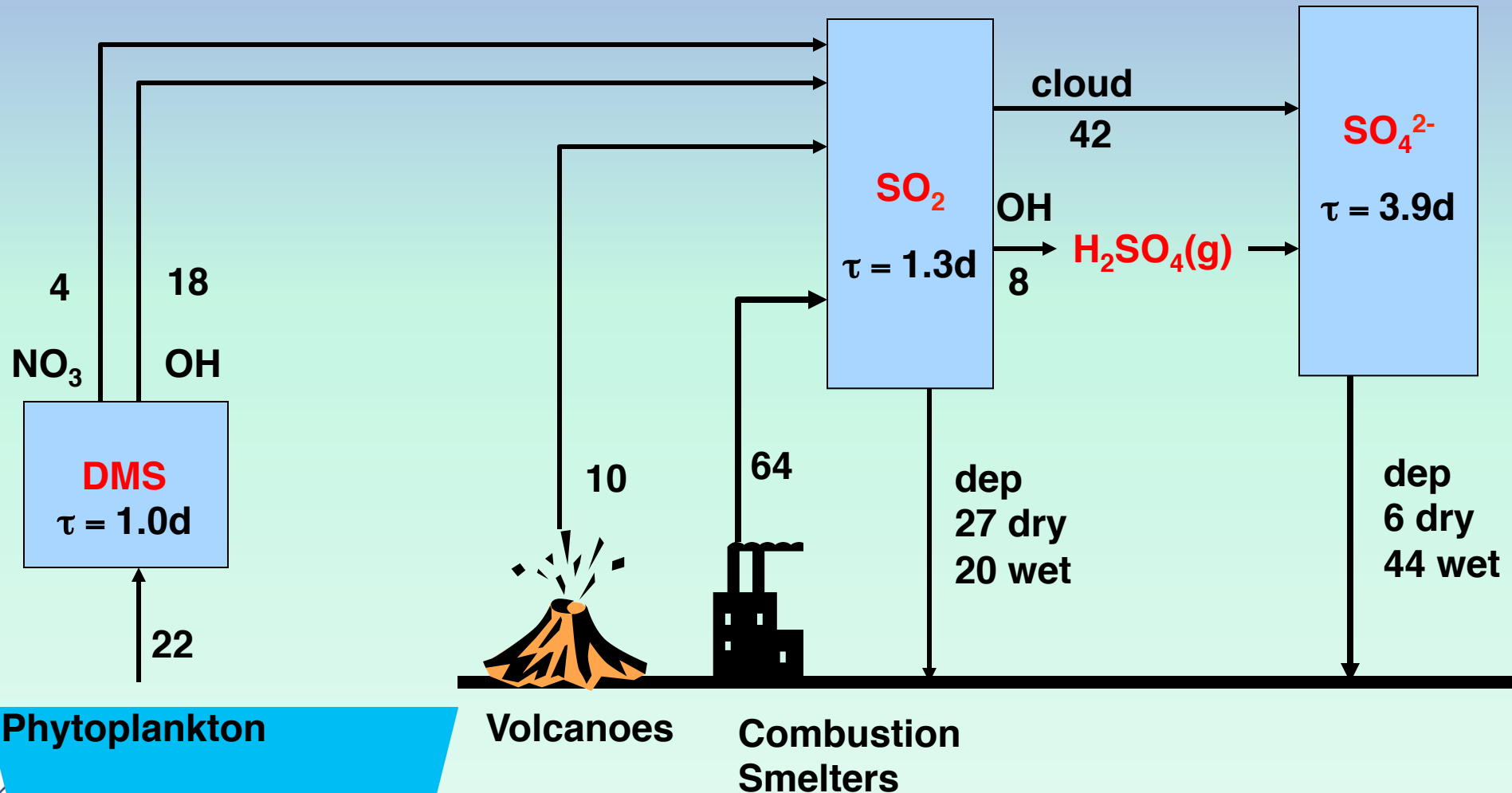
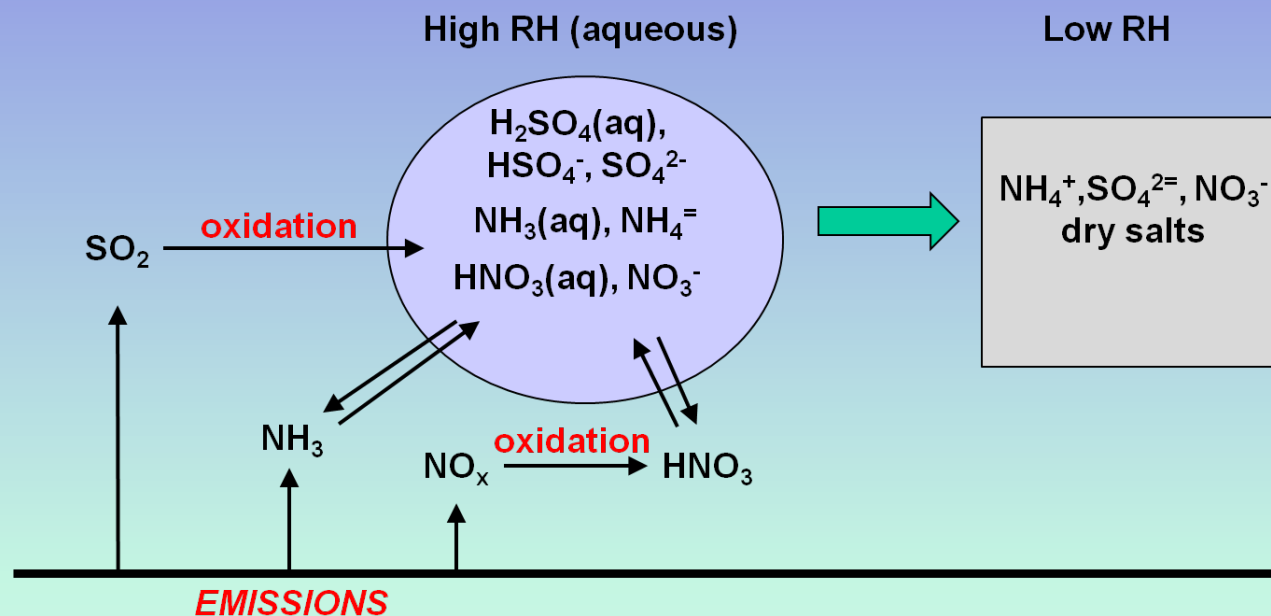


Figure 1. Examples of emission factor size distributions. Upper panel: heavy duty diesel truck emissions for uncontrolled (NOC) and EURO I-VI vehicles. Lower panel: domestic stove emissions for wood (FWD) and hard coal (HC1) fuelled old (NOC), new, improved (IMP), electrostatic precipitator equipped (ESP) and pellet burning (PELL) stoves.

Secondary aerosol: sulfates



Formation of sulfate-nitrate-ammonium (SNA) aerosol



Sulfate always forms an aqueous aerosol

Ammonia dissolves in the sulfate aerosol totally or until titration of acidity, whichever happens first

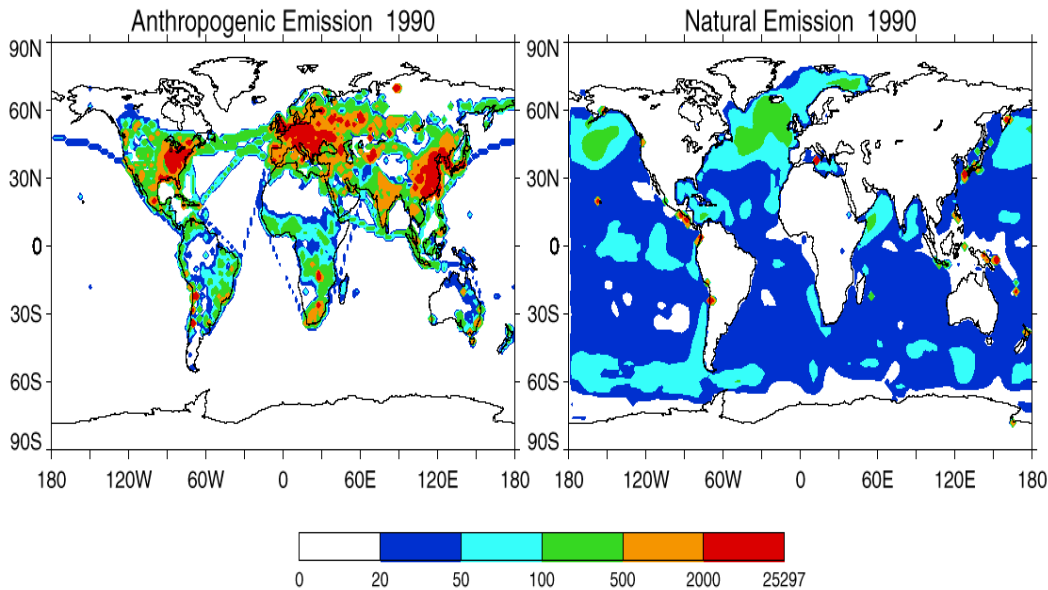
Nitrate is taken up by only if excess NH_3 is available after sulfate titration

HNO_3 and excess NH_3 can also form a solid aerosol if RH is low

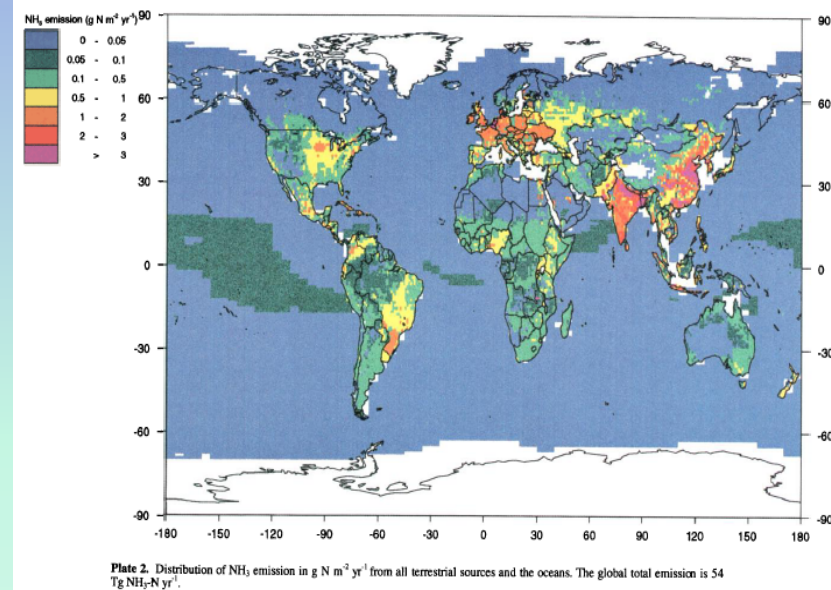
possible effects of cations dissolution if interaction with dust / sea salt ($\text{Ca}^{2+}, \text{K}^+, \text{Na}^+ \dots$)

Global emissions of sulfur and ammonia

SO₂



ammonia

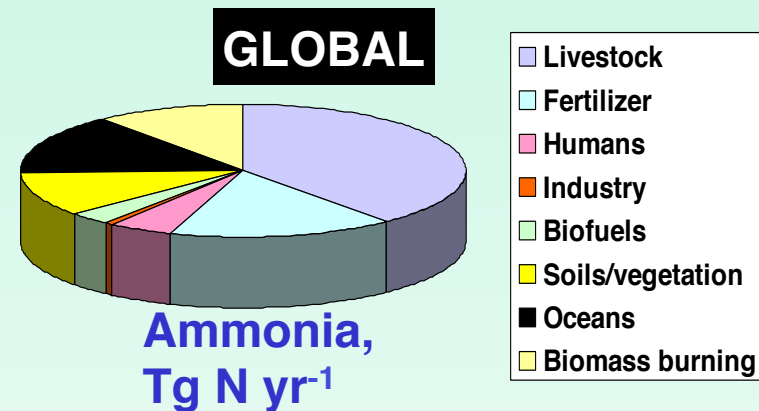


2001 estimates (Tg S yr^{-1}):

Industrial 57 Volcanoes 5

Ocean 15 Biomass burning 1

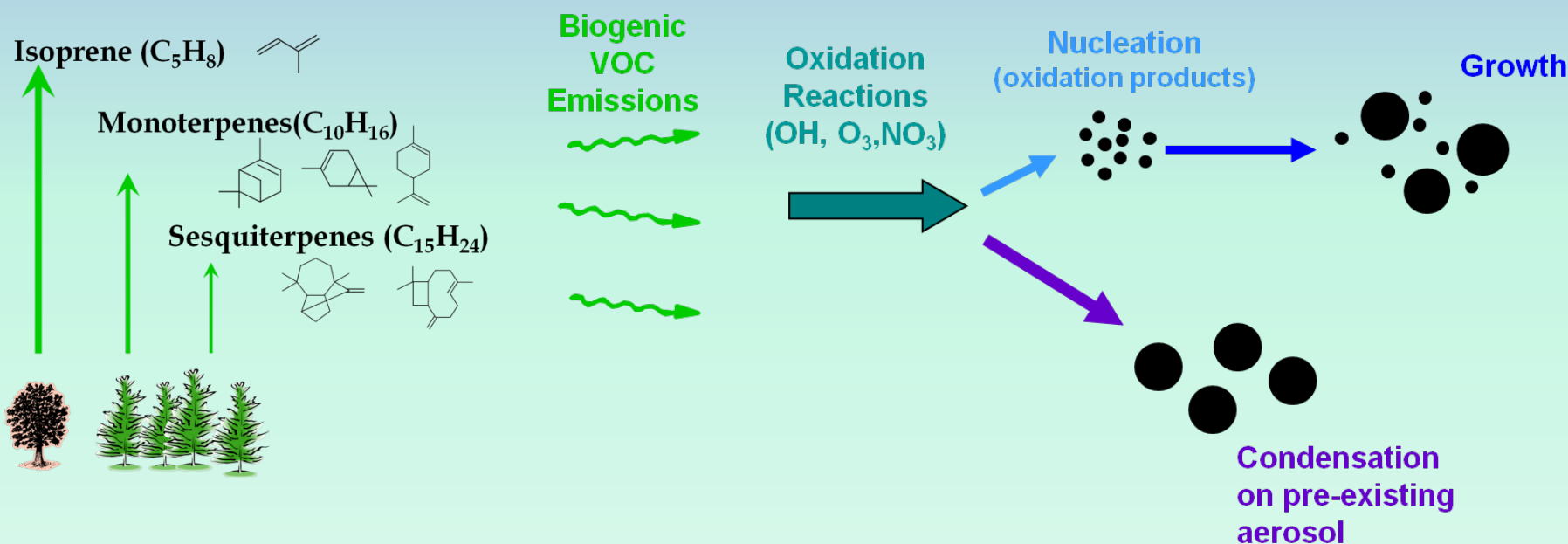
Check out time evolution during practical session



Secondary Organic Aerosol production from biogenic voc emissions

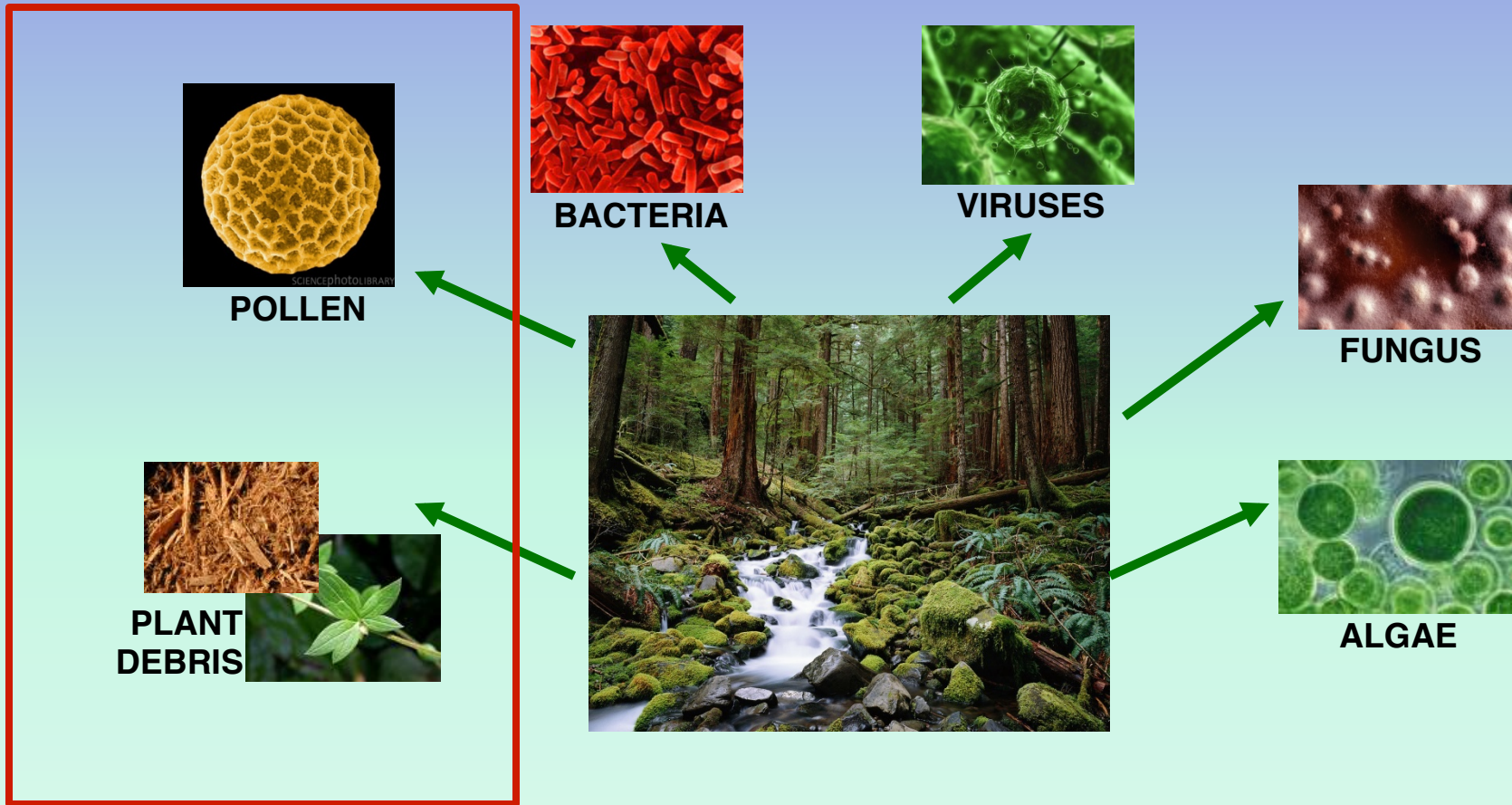
BVOC Production depends on plant species, T, radiation PAR, phenology, water stress (Guenther et al., MEGAN)

Dominate the emission at the global scale



Over 500 reactions to describe the formation of SOA precursors, ozone, and other photochemical pollutants [Griffin et al., 2002; Griffin et al., 2005; Chen and Griffin, 2005]

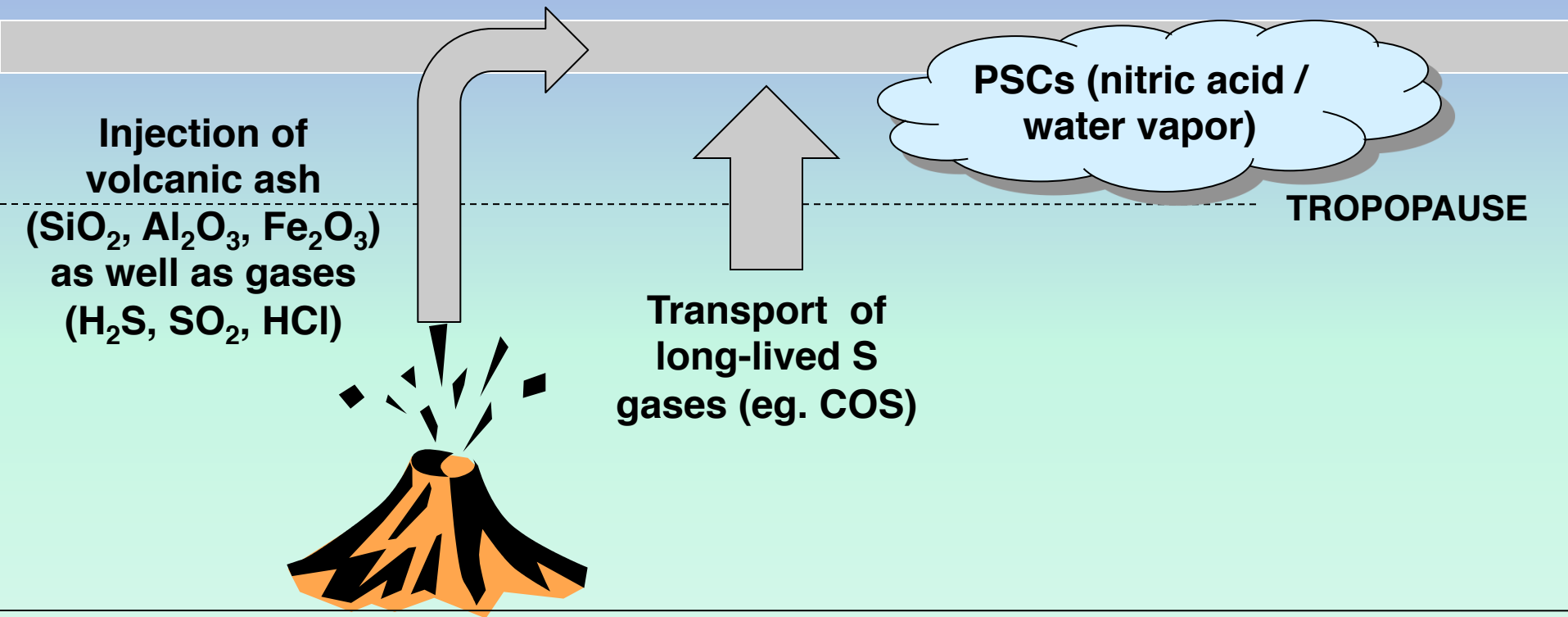
primary biological aerosol particles (pbap)



Very large and likely short-lived

Not much is known / potential for ice nucleation

STRATOSPHERIC AEROSOL



Aerosols in the stratosphere are long-lived due to absence of precipitation and “layered” transport (due to stability)