Aerosol size distributions in atmospheric models

HANNELE KORHONEN

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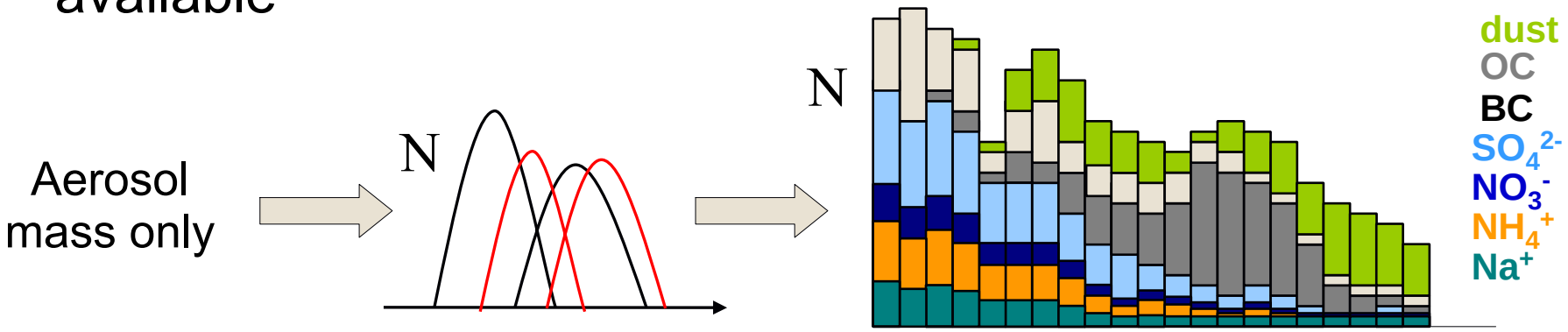
Atmospheric models

- continuous or molecule-by-molecule descriptions impossible to treat in atmospheric models
- description has to be chosen based on aerosol properties of interest + computational resources available



e.g. $PM_{2.5}$ particle number climate impacts

In atmospheric models

- continuous or molecule-by-molecule descriptions impossible to treat in atmospheric models
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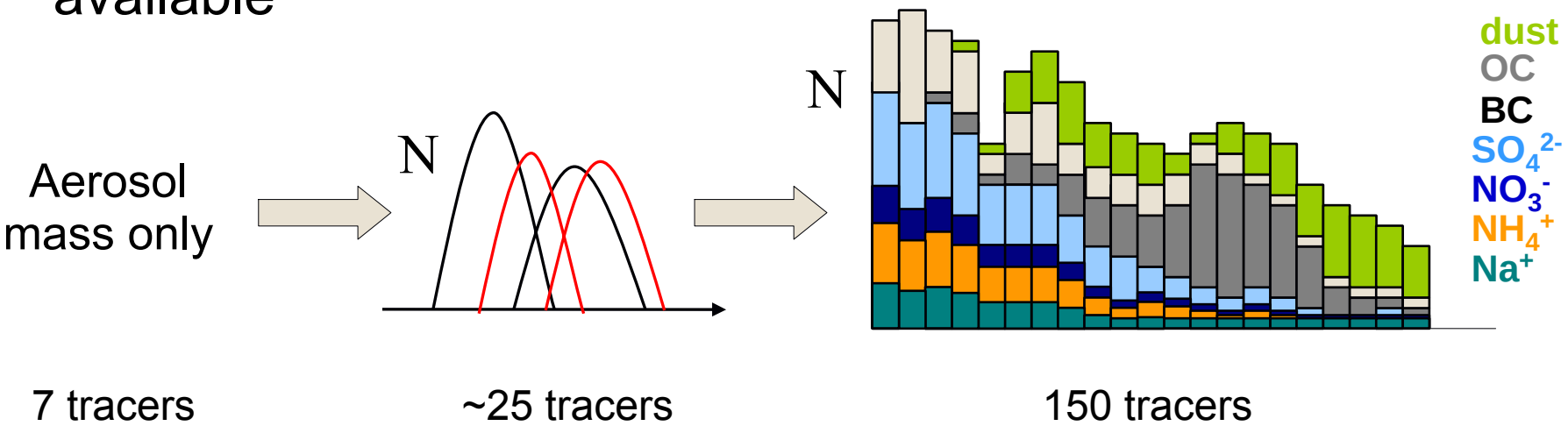


Mass only (bulk) models

– only total mass of aerosol chemical components tracked

dust OC BC SO_4^{2-} NO_3^- NH_4^+ Na^+

–processes not affecting total mass do not need to be described (coagulation, nucleation)

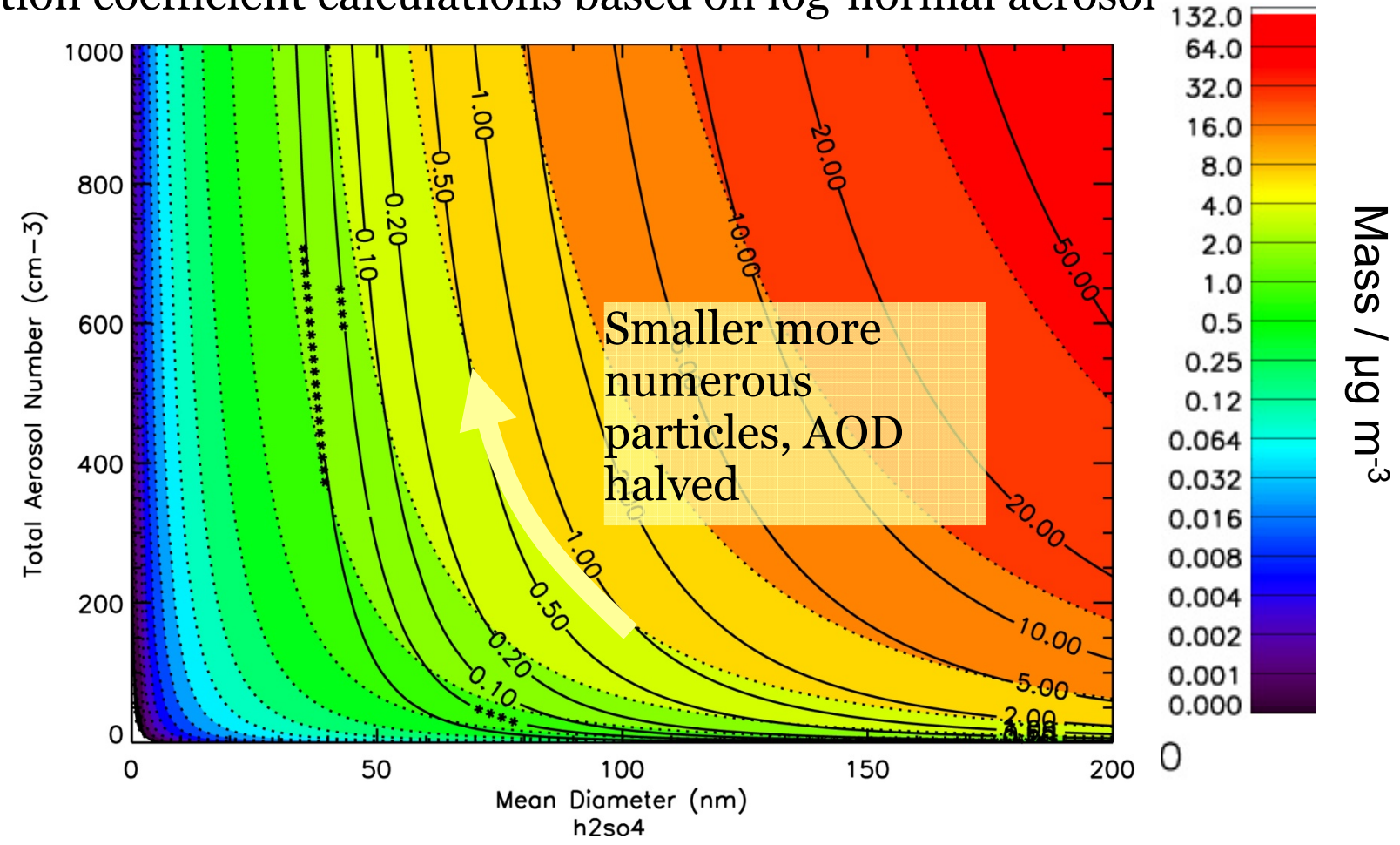
– for size dependent processes (e.g. deposition, climate effects)

1. "typical" size distribution assumed
2. process parameterized based on measurements

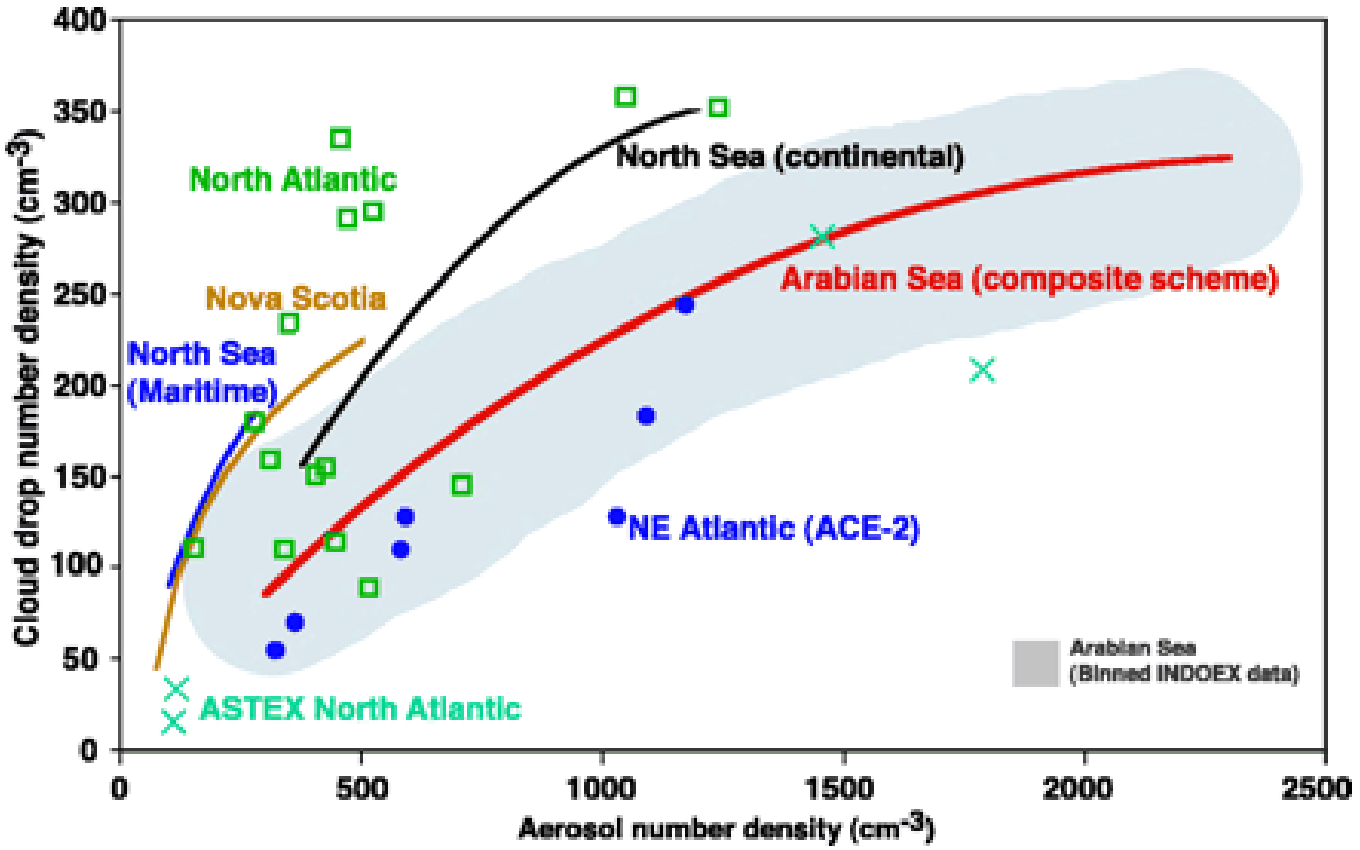
   e.g. $\log(CDN) = 2.38 + 0.49\log(M_{SO4})$

Problems with"typical" distributions

Extinction coefficient calculations based on log-normal aerosol



Problems with measurement-based parameterisations



Equilibration (instead of condensation)

Mass only models often use equilibrium assumptions

- direct minimisation of Gibbs free energy
- solving a set of non-linear equations from mass balances and chemical equilibrium

Several equilibrium models available especially for inorganic species (SO_4, NO_3, NH_4, Na, Cl, Ca, Mg)

- e.g. ISORROPIA, EQUISOLV II, EQSAM,...
- input: temperature, RH, total concentration of species in the atmosphere (gas + aerosol phase)
- output: equilibrium concentration of species in gas and aerosol phases at given conditions

Equilibration: organics

For organics in bulk models, Pankow approach is common.

Assume that each organic precursor reacts to form 2 condensable compounds (α_i is stoichiometric coefficient of *i*)

$$P \rightarrow \alpha_1 C_1 + \alpha_2 C_2$$

Each of these compounds partitions according to

$$K_{p,om,i} = \frac{(ng/\mu g)_{om\ phase}}{(ng/m^3)_{gas\ phase}} = \frac{F_i/M_o}{A_i} = \frac{760RT}{10^6\ MW_{om}\zeta_i p^\circ_{L,i}}$$

where M_0 is mass of formed aerosol, A_i concentration in gas phase and F_i concentration in organic aerosol phase.

Equilibration: organics

Note that equation

$$K_{\mathrm{p,om},i} = \frac{(\mathrm{ng}/\mu\mathrm{g})_{\mathrm{om\ phase}}}{(\mathrm{ng}/\mathrm{m}^3)_{\mathrm{gas\ phase}}} = \frac{F_i/M_{\mathrm{o}}}{A_i} = \frac{760RT}{10^6\ \mathrm{MW}_{\mathrm{om}}\zeta_i p^{\circ}_{\mathrm{L},i}}$$

implies that SOA formation rate is dependent on the amount of organic material *already present* in aerosol phase!

$K_{p,om,i}$ values are determined from laboratory experiments.

Pros and cons of mass only approach

Pros

– computationally very fast

–typically sufficient for air quality models interested only in PM_{10} or $PM_{2.5}$

Cons

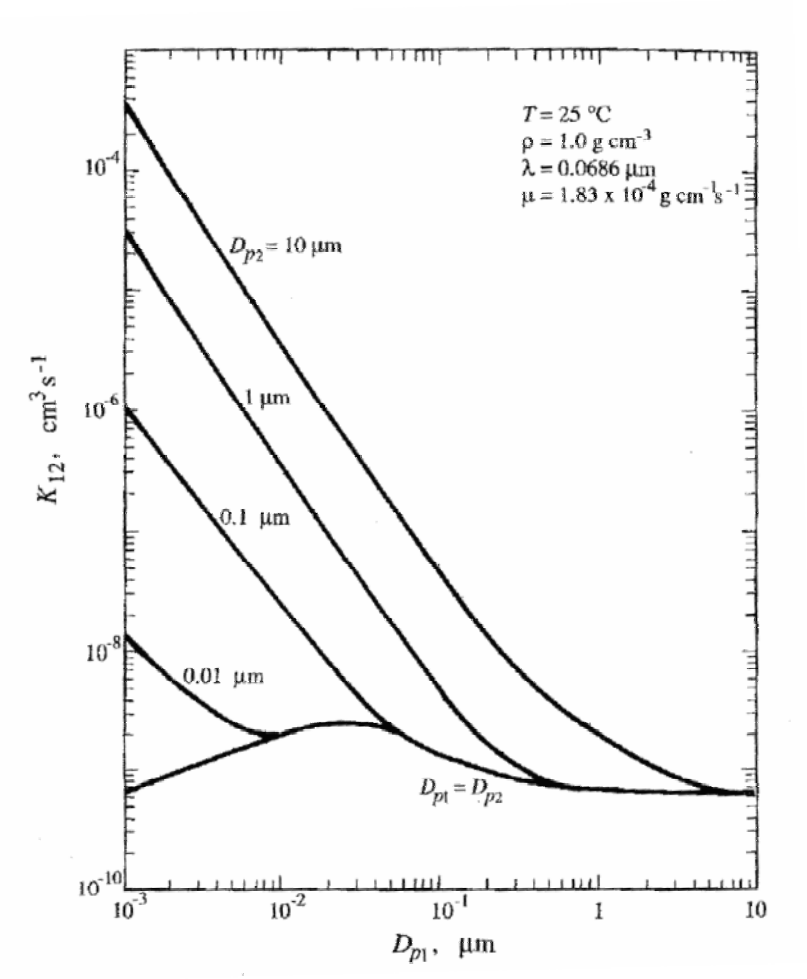
– cannot be used in number concentration or climate studies

–may be unrealiable if wet deposition (size dependent!) is important

Size-segregated approaches

Basically all aerosol processes are size dependent.

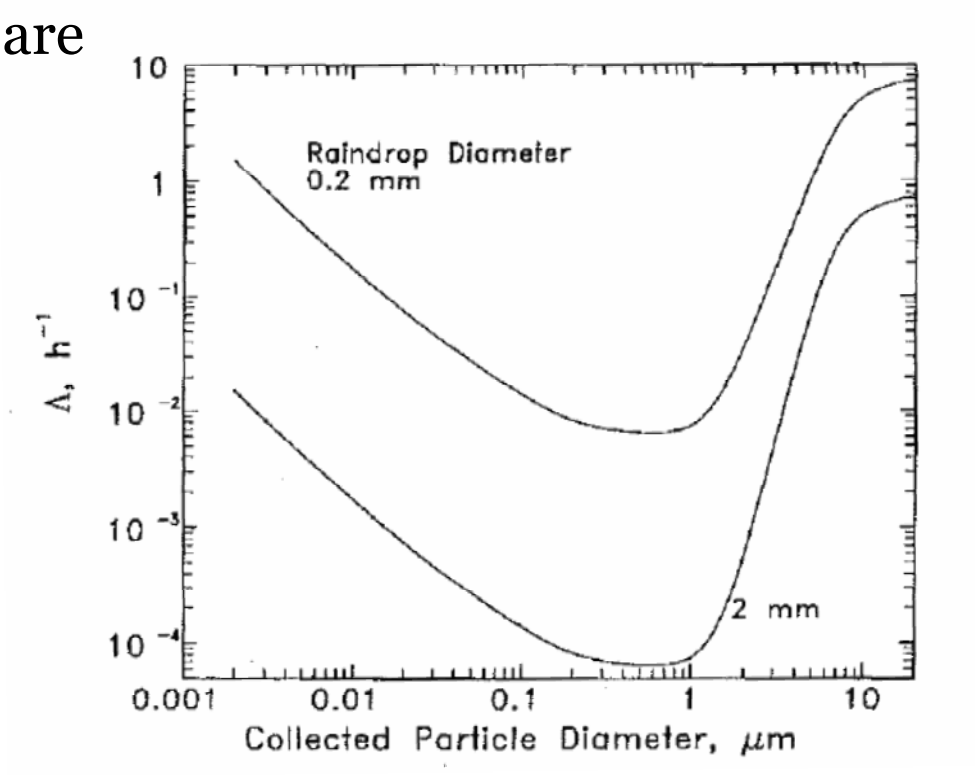
E.g. coagulation coefficients



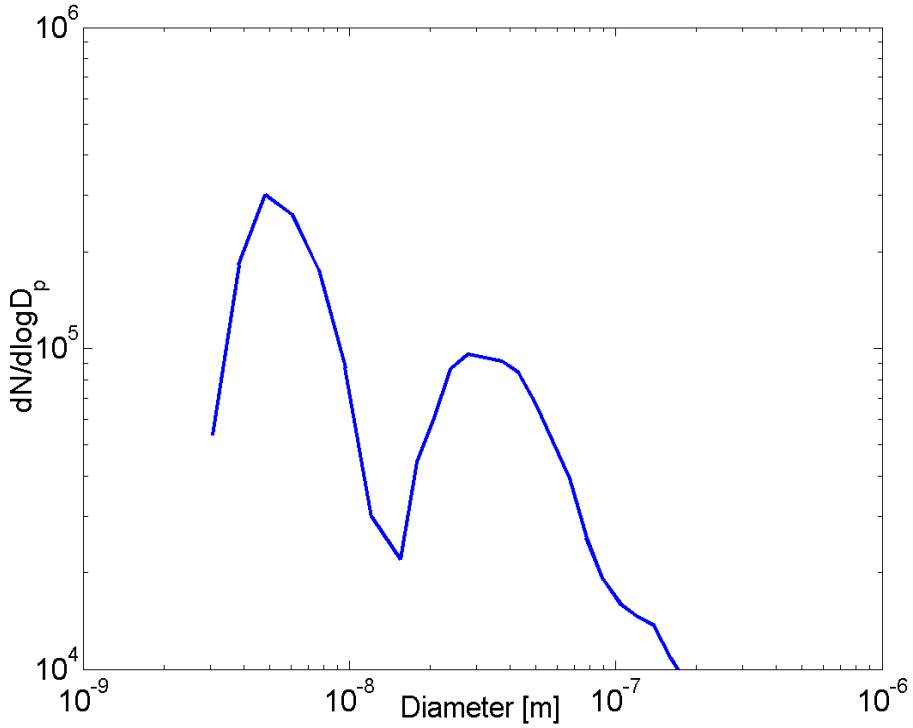
Size-segregated approaches

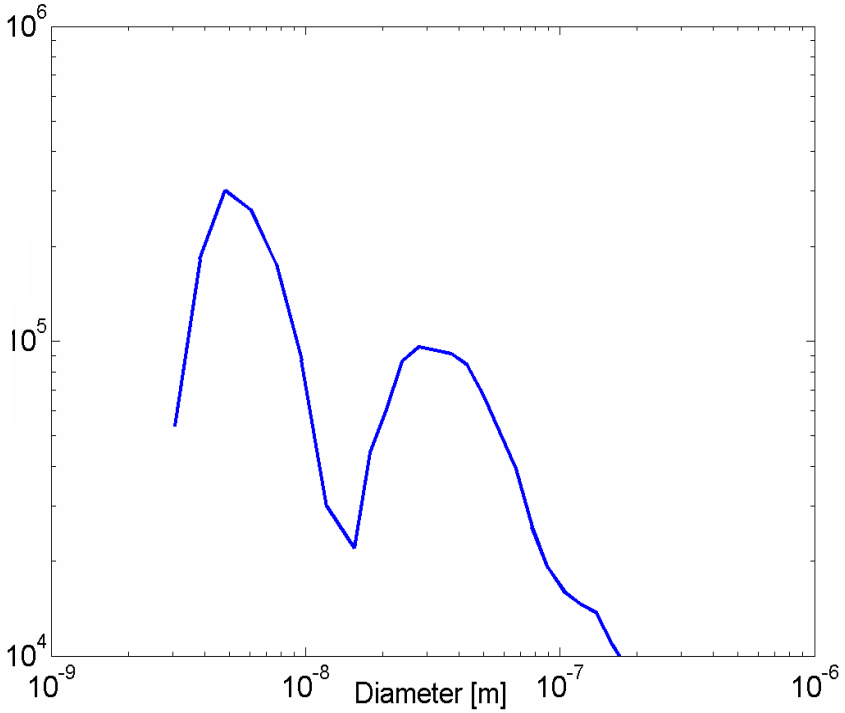
Basically all aerosol processes are size dependent.

E.g. impaction scavenging (below- cloud wet deposition)

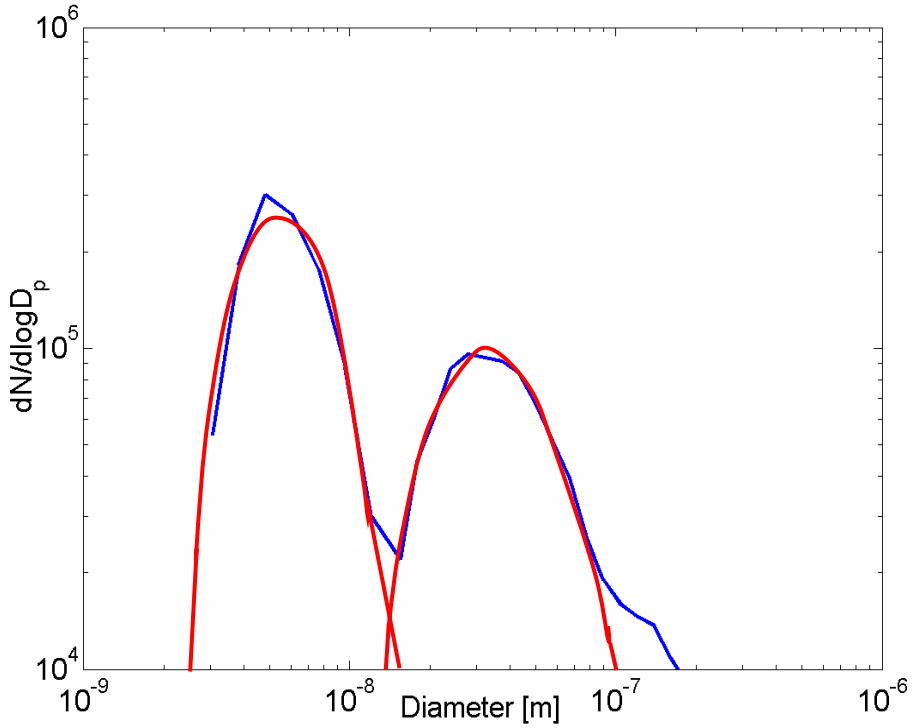


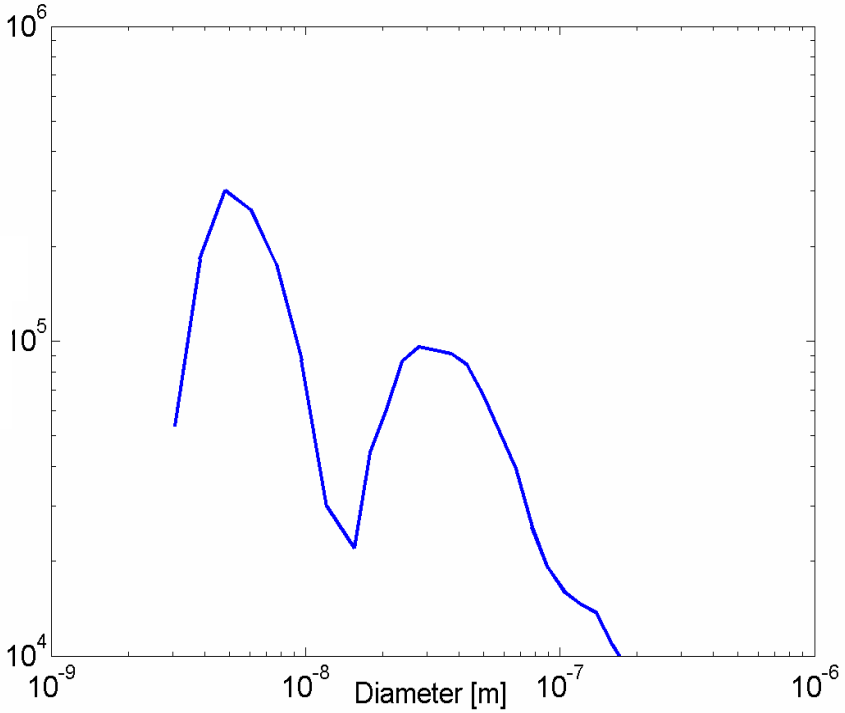
Size-segregated approaches





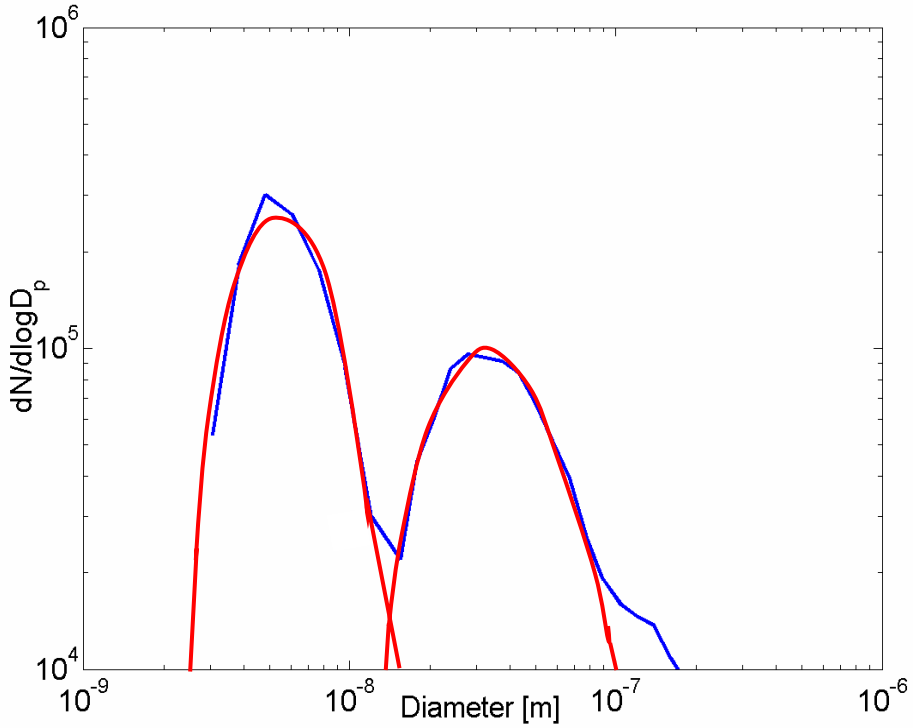
Size-segregated approaches



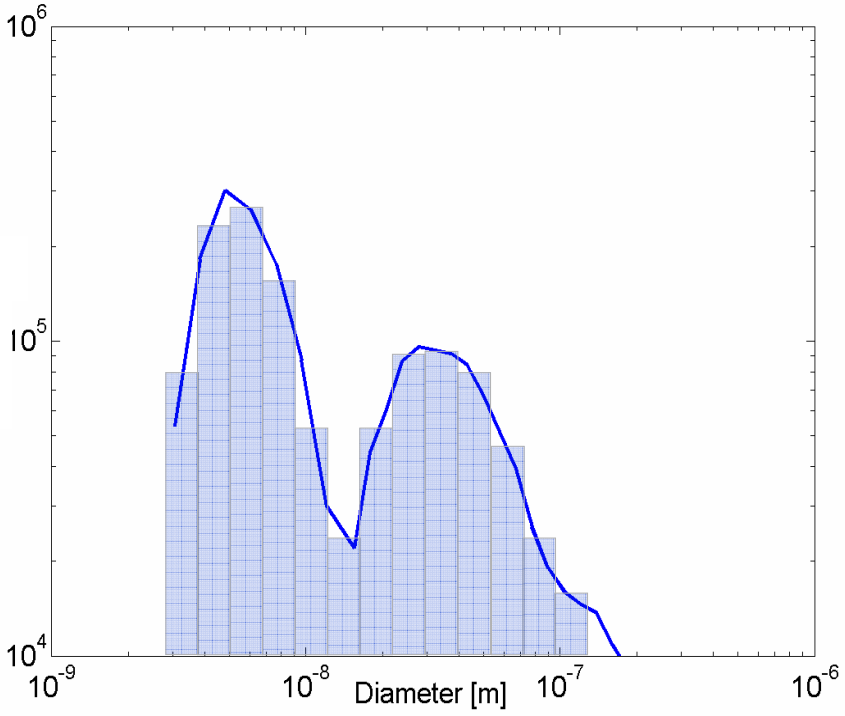


Modal approach

Size-segregated approaches

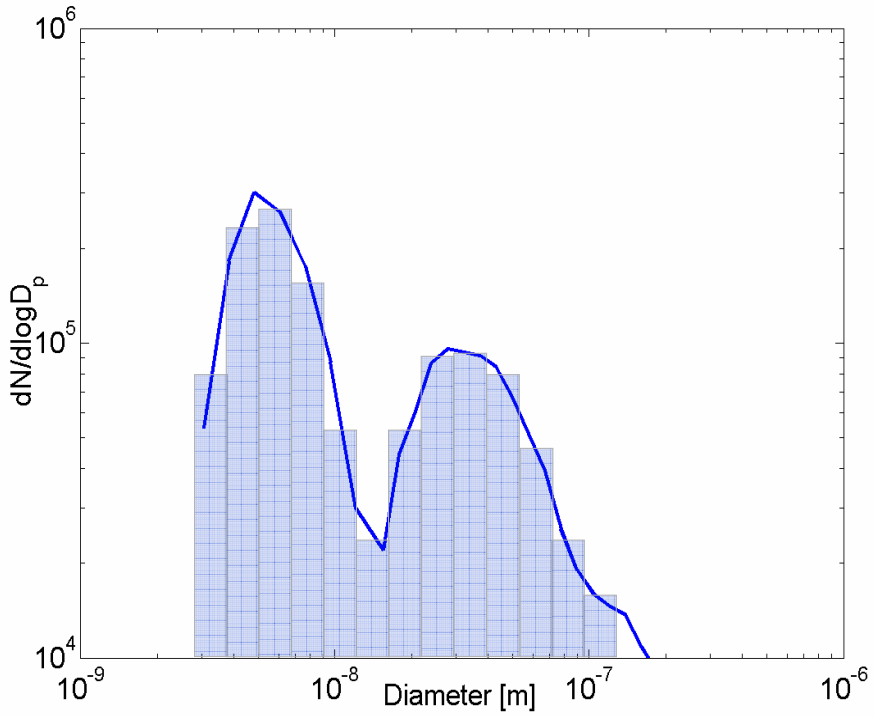


Modal approach



Sectional approach

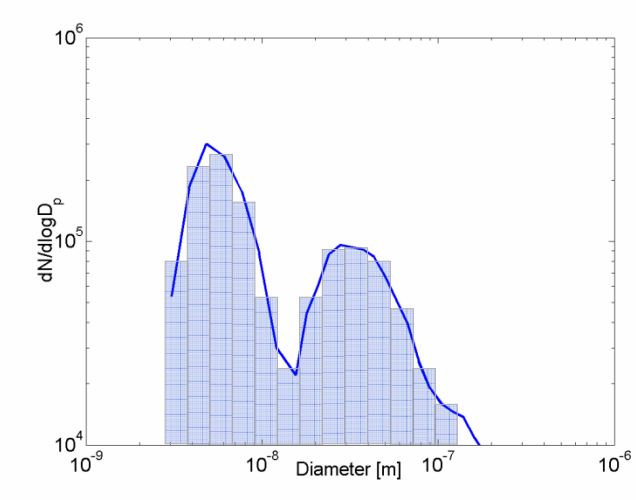
Sectional approach

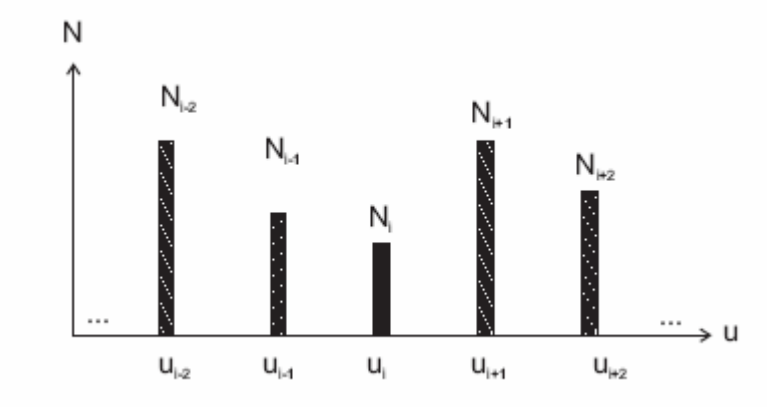


Number of size sections

- ~10 to several hundreds
- strongly affects accuracy
- affects computational burden
  - n sections, m components
    → n x m differential eqns

Sectional approach





- size distribution function becomes

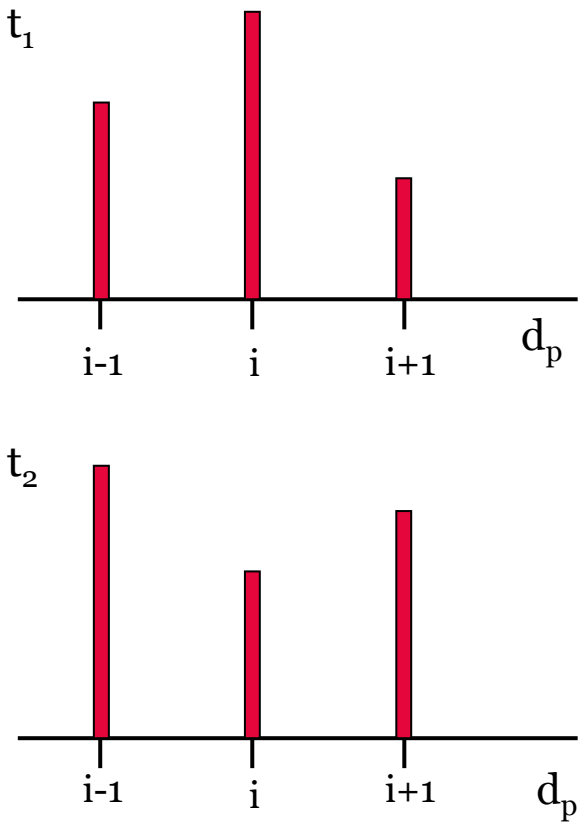
$$n(d_p) = \frac{dN}{d(log(d_p))} \sim \frac{\Delta N}{log(d_{max}/d_{min})}$$

  i.e. flat distribution within size sections

- often size distribution *within* size sections is assumed monodisperse

- in both cases, size sections are typically spaced logarithmically

I. Fixed size sections



- sections have fixed locations in size space
- e.g. condensation moves particles to larger sections
- original (and still commonly used) sectional formulation

I. Fixed size sections

Advantages

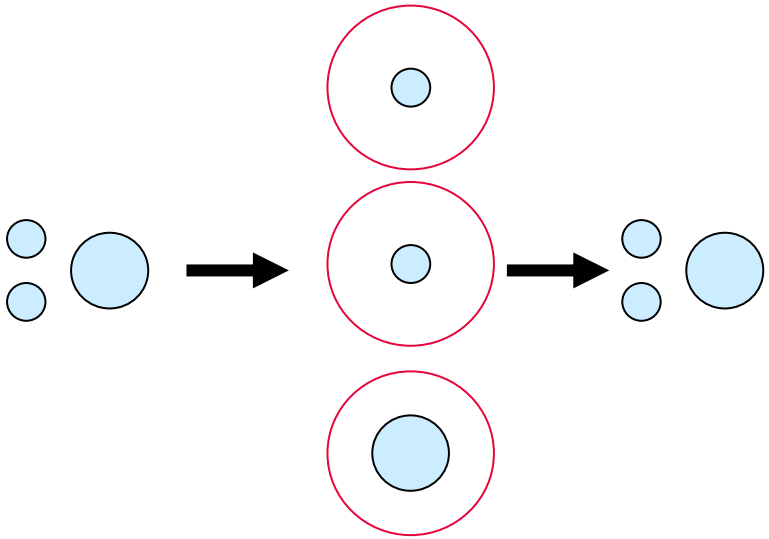
- easy to code
- accurate treatment of coagulation (usually!), nucleation, deposition
- easy treatment of transport in 3D models

Disadvantages

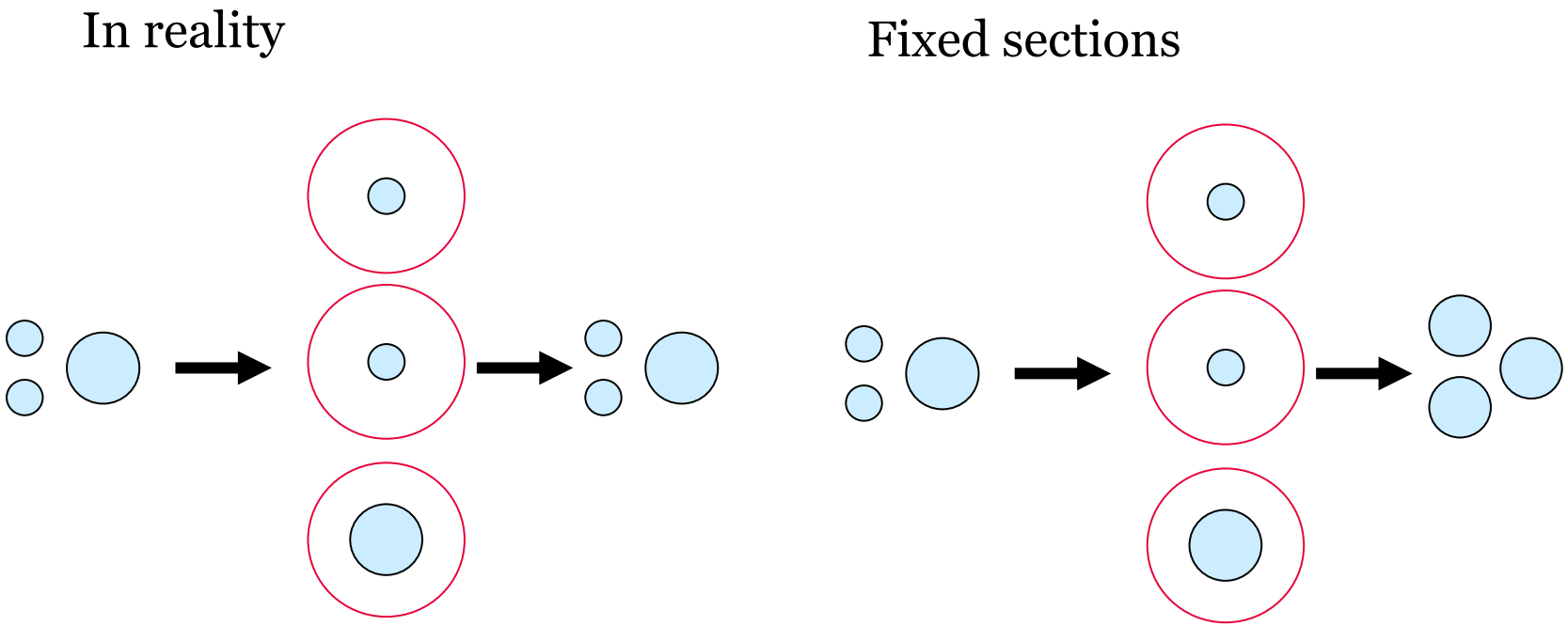
- loss of resolution upon growth (important e.g. in cloud applications)
- treatment of condensation produces numerical diffusion

Loss of resolution upon growth

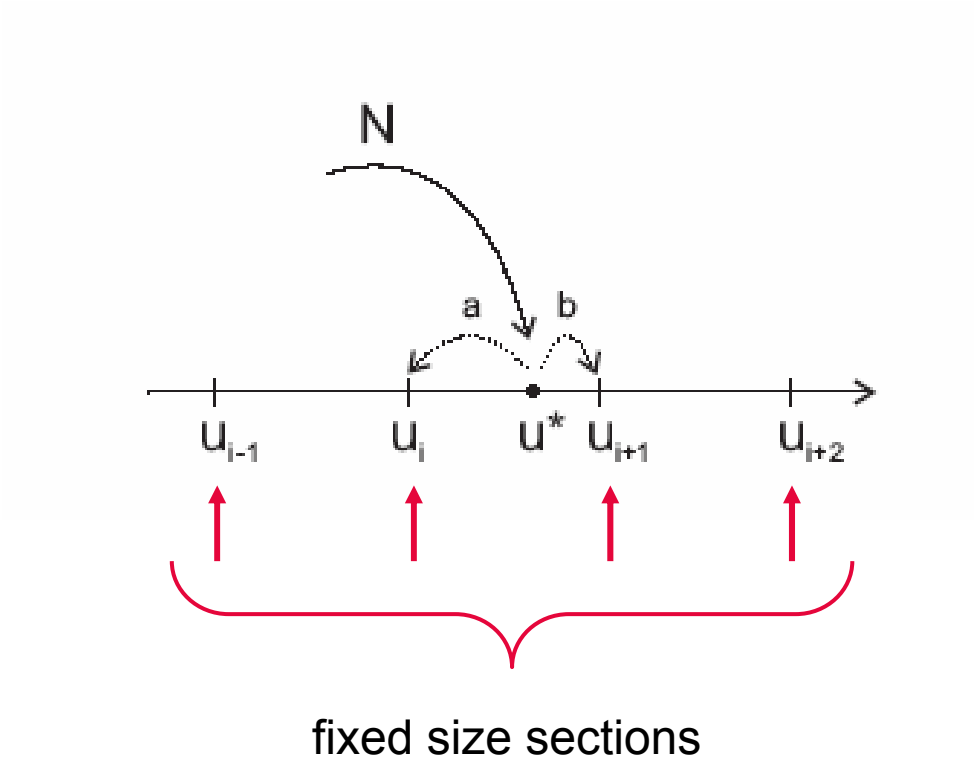
In reality



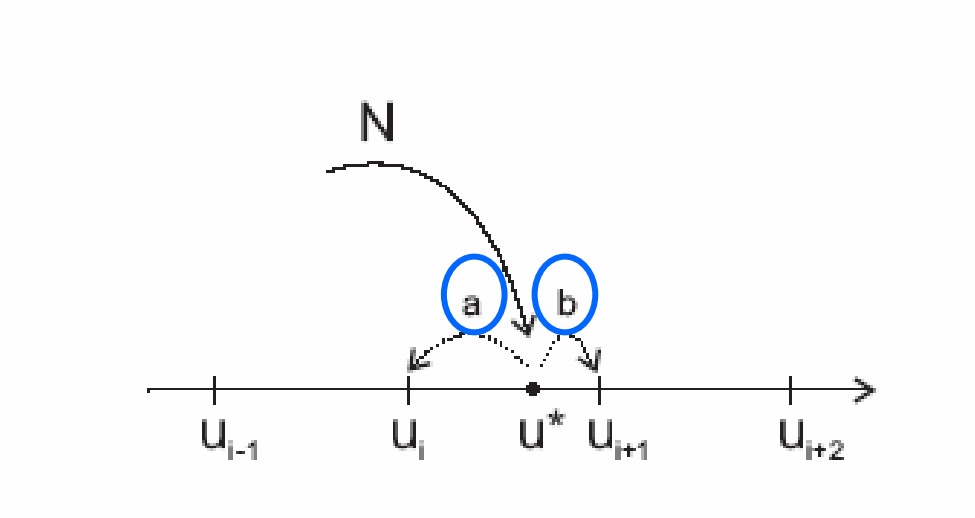
Loss of resolution upon growth



Size splitting: the concept



Size splitting: the concept



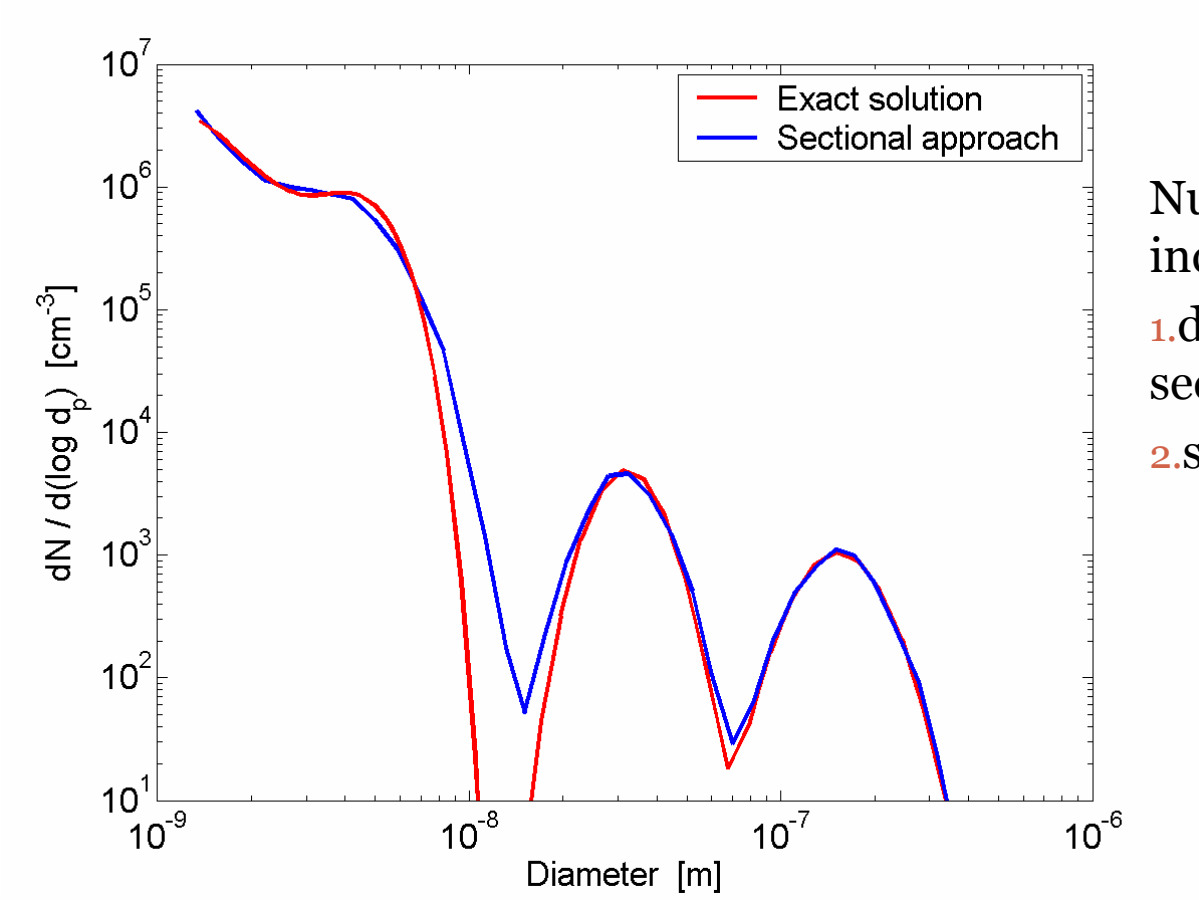
Conserve particle number *and* volume

$$\begin{cases} a + b & = & N \\ a u_i + b u_{i+1} & = & N u^\star \end{cases}$$

$$\Rightarrow$$

$$\begin{cases} a & = & \frac{u_{i+1} - u^*}{u_{i+1} - u_i} N \\ b & = & \frac{u^* - u_i}{u_{i+1} - u_i} N \end{cases}$$

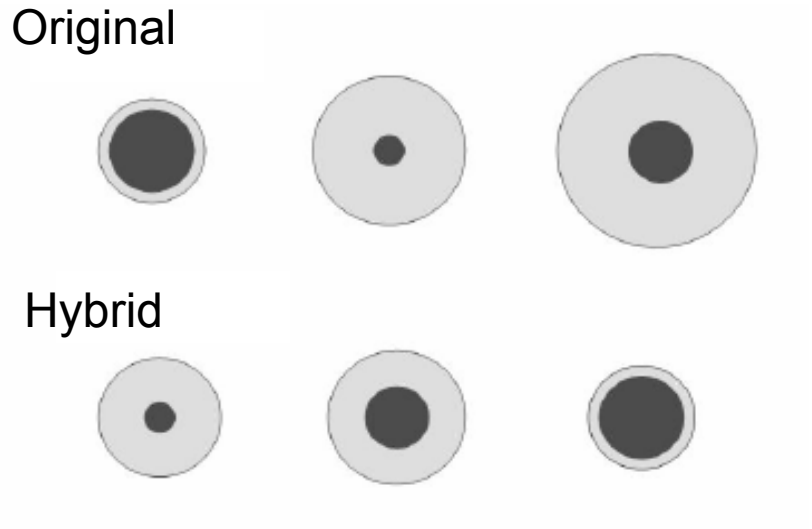
Size splitting causes numerical diffusion



Numerical diffusion increases with

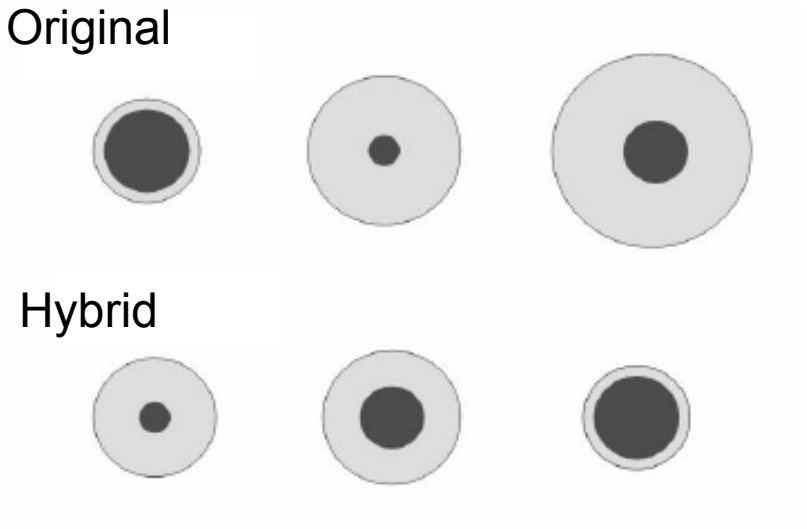
1. decreasing number of size sections
2. shortening of time step

Ib. Modified fixed method: hybrid approach



- Particle volume divided into
  - nonvolatile core
  - volatile surrounding layer
- Fixed sections according to core (*not total!*) volume

Ib. Modified fixed method: hybrid approach



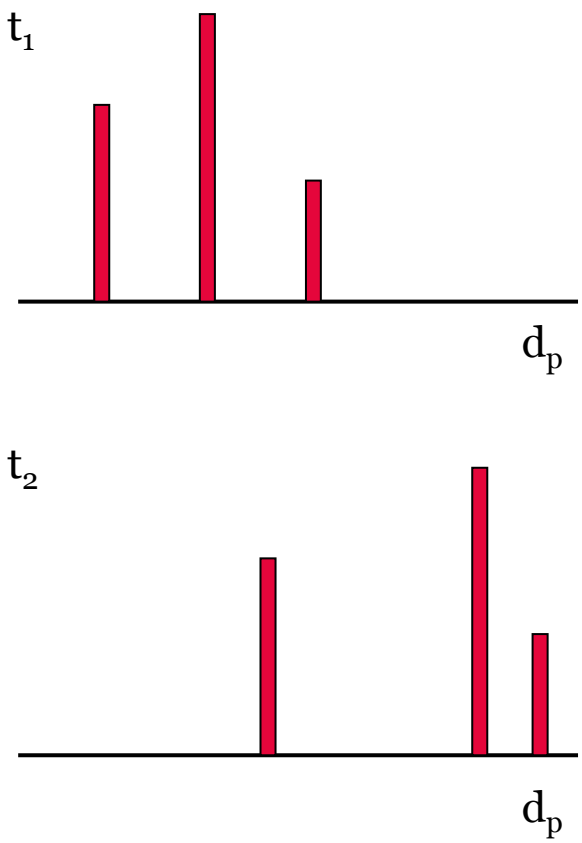
Advantages

- numerical diffusion only when core compounds condense

Disadvantages

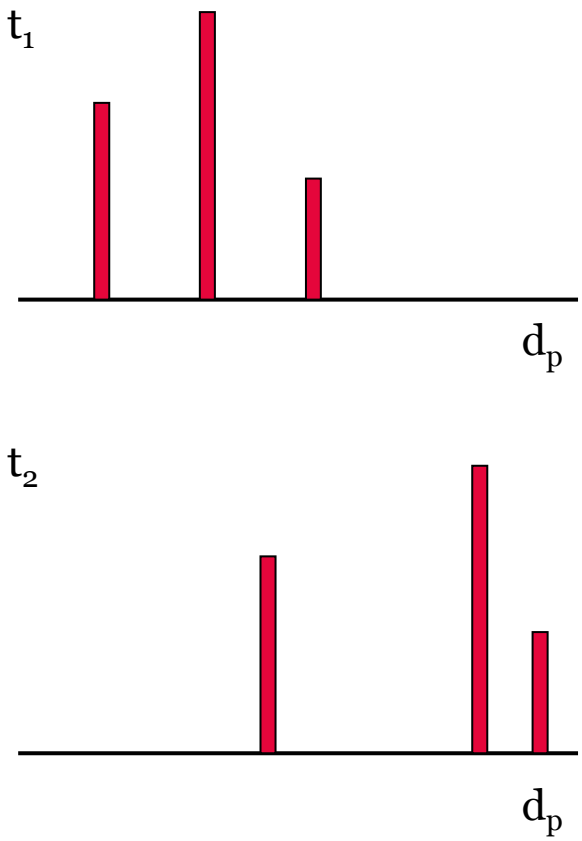
- need to recalculate ambient size of particles (and thus e.g. coagulation coefficients) at all time steps

II. Moving size sections



- Size sections move in size space according to particle growth
- In principle, the order of the sections can change during the simulation
- Advantages
  - no numerical diffusion or loss of resolution
  - very easy to program

II. Moving size sections

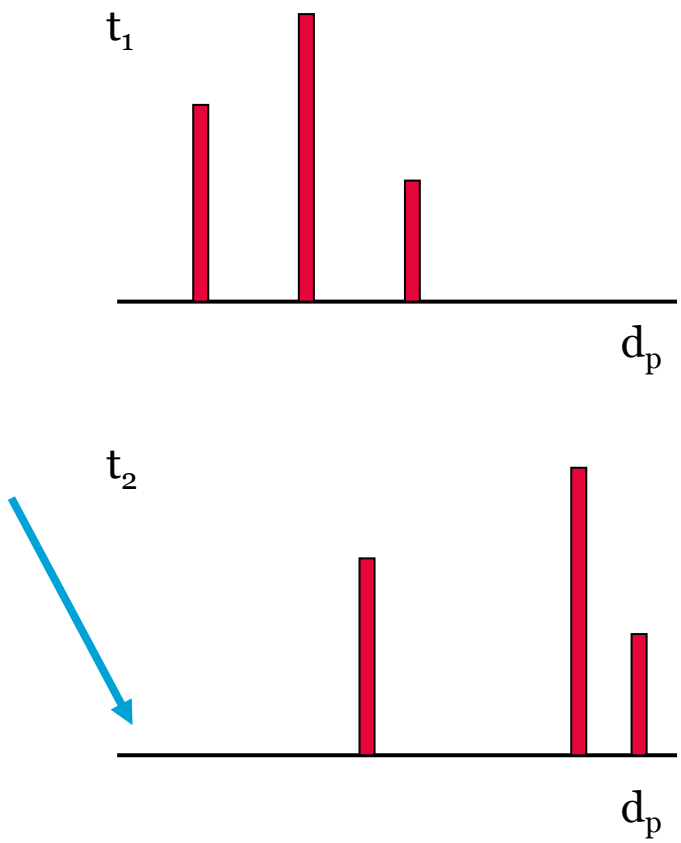


Would there be problems with

1. nucleation
2. coagulation
3. transport?

If so, any suggestions how to solve them?

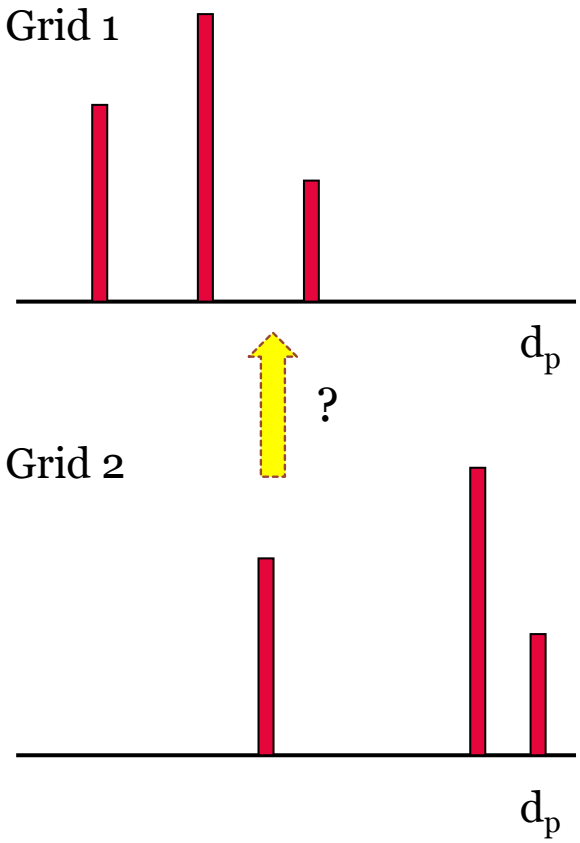
Moving size sections: nucleation



Two possibilities

1. "pull back" the smallest bin (not very realistic)
2. create new bins for forming particles (accurate but can become computationally heavy)

Moving size sections: transport

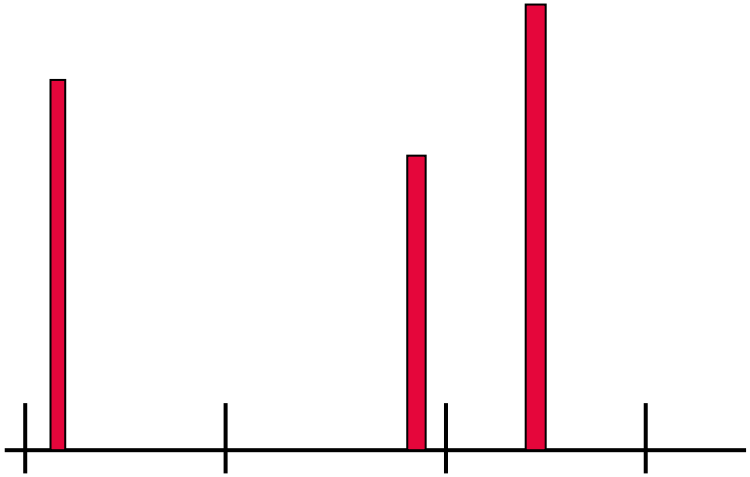


Bins in adjacent grids are not (necessarily) at same sizes.

In practice, moving grid needs to be retracked into a fixed grid at every transport time step

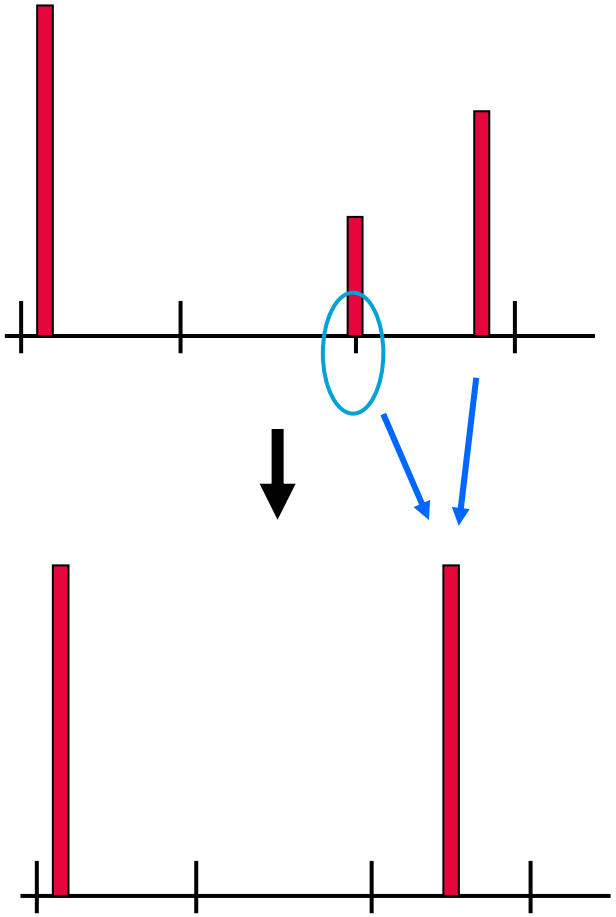
→ numerical diffusion

III. Moving centre method



- Section boundaries fixed
- Particle size can vary within the section

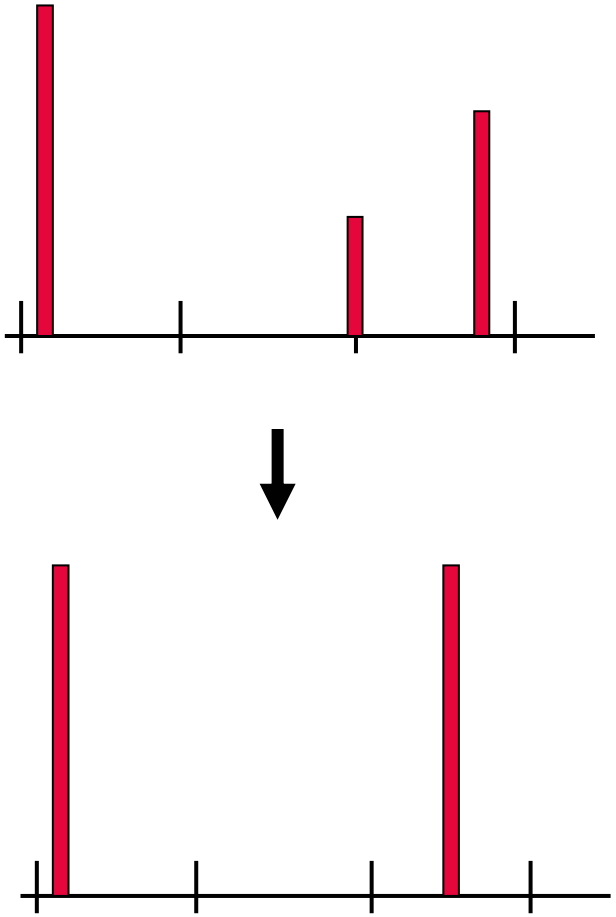
III. Moving centre method



- when boundary reached
  → all particles moved to adjacent section

- new characteristic size of section by averaging (number and volume conserved)

III. Moving centre method



Advantages

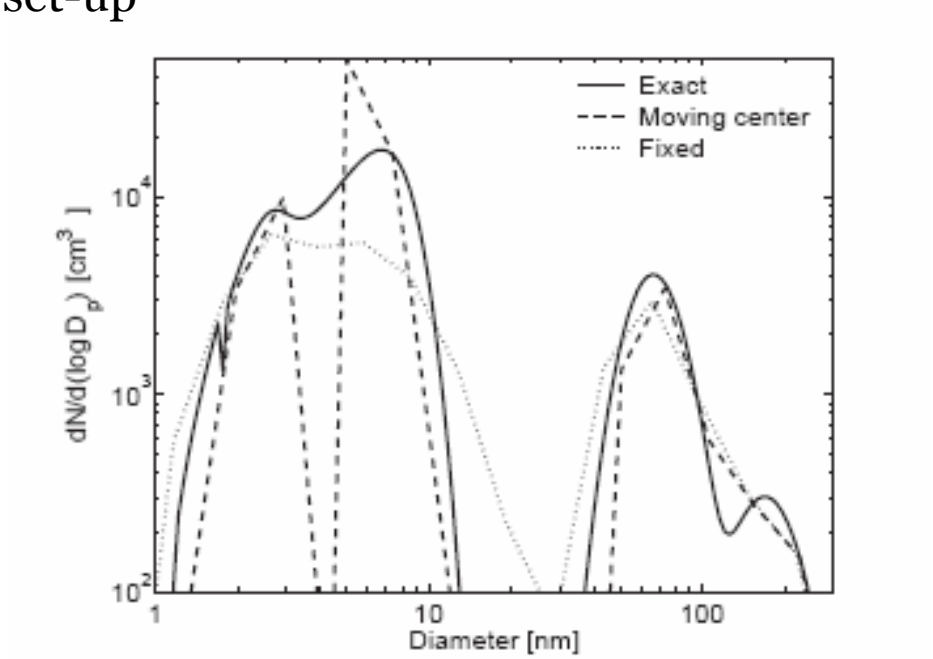
- little numerical diffusion
- nucleation, emissions, transport realistically treated

Disadvantages

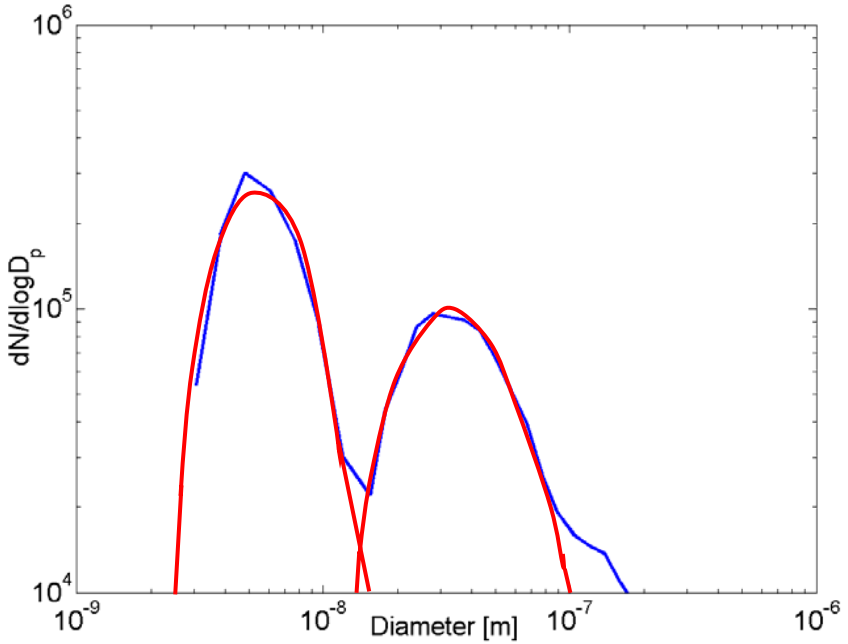
- loss of resolution upon growth
- empty bins (in 3D models smoothed out)
- e.g. following new particle growth tricky because particles ”pulled back” when averaged

Comparison of sectional approaches

Box model set-up

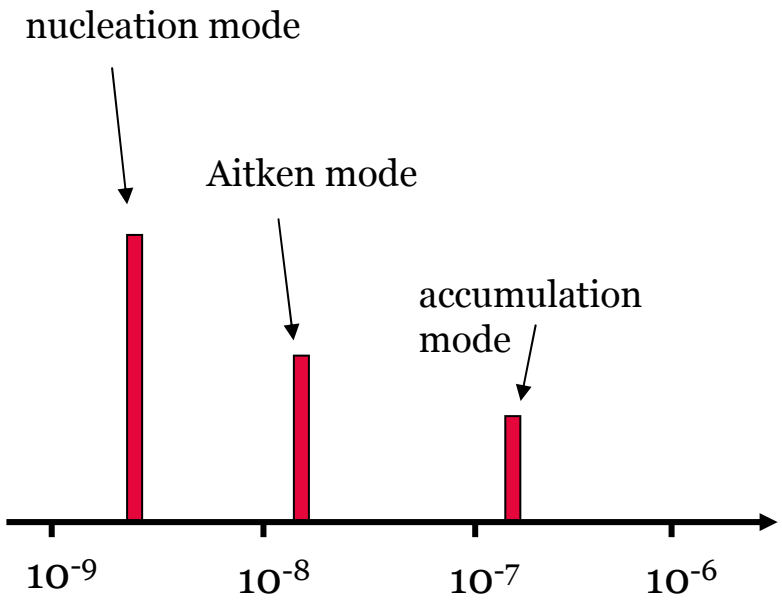


Modal approach



- Size distribution described with modes (typically 2+)
- Modes move in the size space according to particle growth
- Assumptions about the shape of the mode must be made

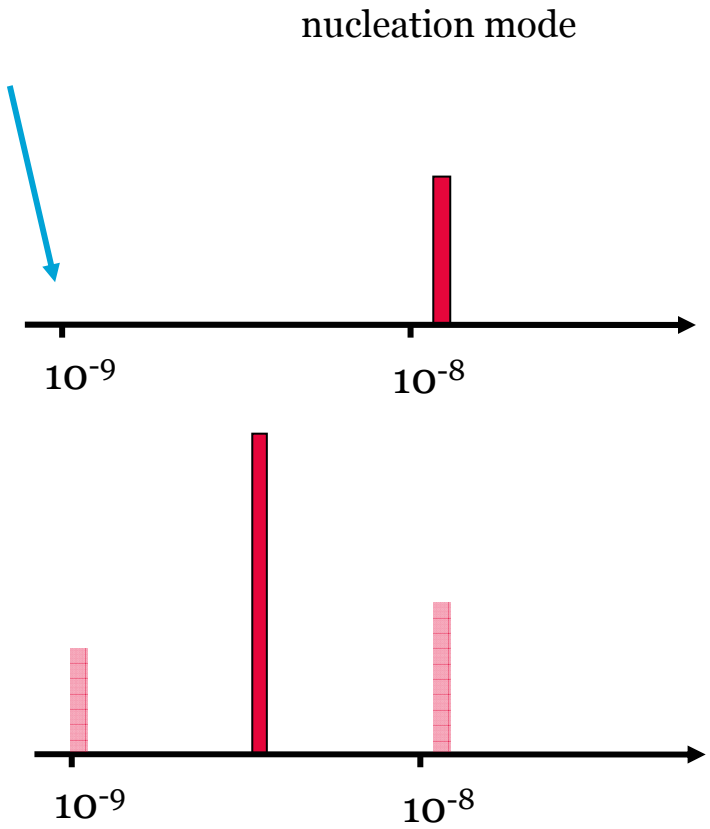
I. Multimodal monodisperse approach



Assumptions

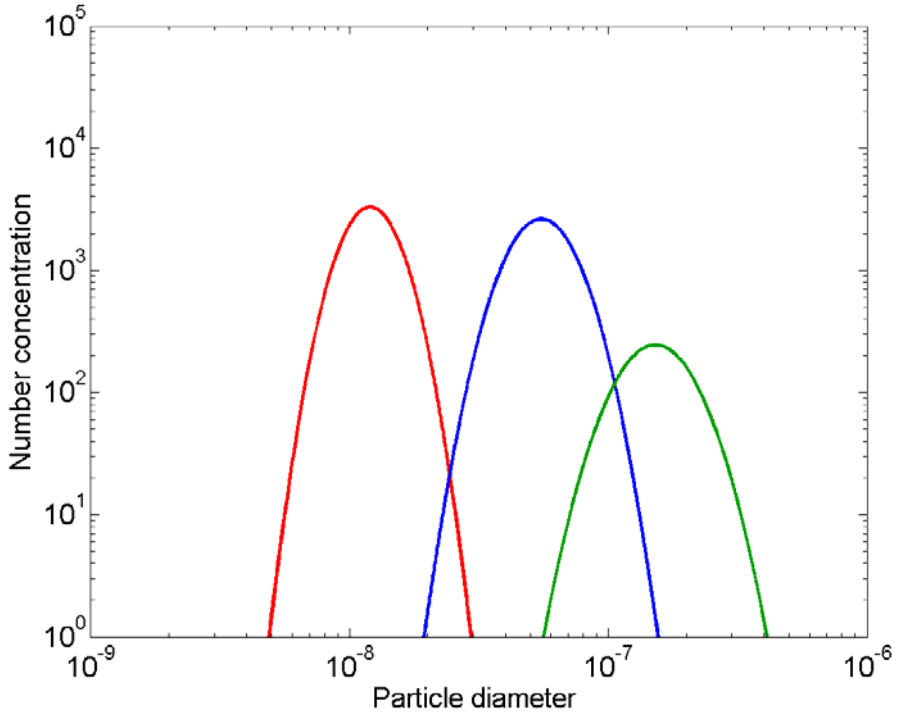
- condensation moves modes to larger sizes
- nucleated particles to smallest mode
- when coagulating, particles placed into mode of larger original particle

I. Multimodal monodisperse approach



- Advantages
  - computationally fast
  - easy to code
- Disadvantages
  - very crude representation
  - treatment of nucleation!!!
  - problems with emissions and transport

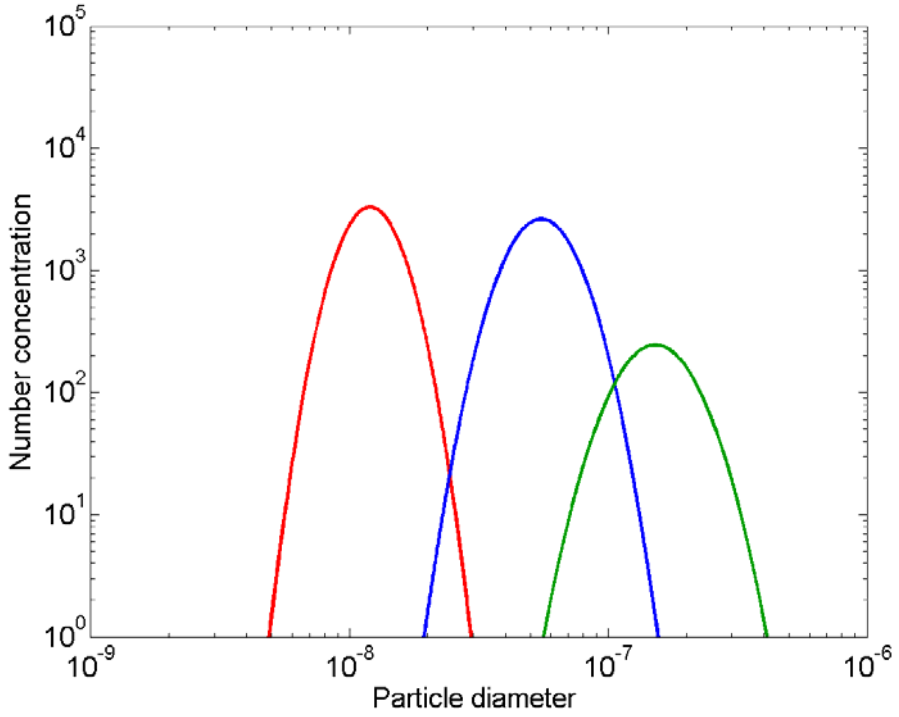
II. Log-normal approach



- Assumes that each mode is log-normal in shape
  - fairly realistic
  - modes can be represented with (2 or) 3 variables
- Tracks low-order radial moments of modes

$$M_k = \int d_p^k n(d_p) dd_p$$

II. Log-normal approach



0th moment gives N

$$M_0 = \int n(d_p) dd_p$$

- 3rd moment proportional to V

$$M_3 = \int d_p^3 n(d_p) dd_p$$

- e.g. width of the mode from a combination of the moments

II. Log-normal approach

Advantages

- computationally efficient → popular in e.g. regional air-quality models
- internal/external mixtures can be treated easily

Disadvantages

- prescribed shape of distribution (not always realistic)
- difficult to treat step functions (e.g. cloud activation)
- potentially problems with nucleation, emissions, transport

Summary

- If you are interested only in aerosol mass, go with bulk approach
  - NB: careful with parameterisations and "typical" distributions
- When size information is needed, first choice in large scale models is modal log-normal approach
  - Most recent versions of approach show fairly good agreement with mass, CCN and even size distribution measurements
- Most detail is achieved with moving centre sectional approach but it is computationally expensive to run
  - for detailed simulations of climate effects, for aerosol CTMs…