Modelling of externally mixed particles in the atmosphere

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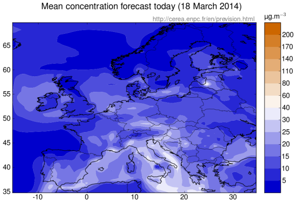
- Aerosols: impact on both human health and climate
- Air quality models to forecast and study air pollution
- Internally mixed aerosol model: *single composition for one particle size*
  - Assumption: *particles of the same size mix instantaneously when they meet*
- Externally mixed aerosol model: *Multiple compositions for each particle size*

  Advantage:
  - *Retain the source identity of particles*
  - *Enable to analyse the particle mixing state (Important: radiative forcing; hygroscopic character; etc)*

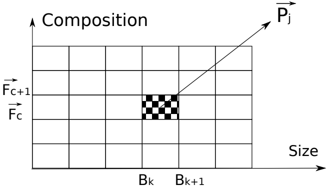
Paris, March 18, 2014



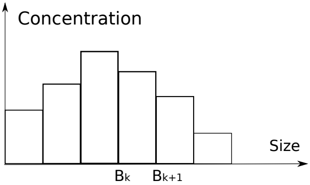
$PM_{2.5}$ forecast (CEREA)

Model description

- SCRAM: Size-Composition Resolved Aerosol Model
- Discretization: $\vec{P_j} = (\vec{F_c}, d_k)$
  - Particle diameter: $d_k \in [B_k, B_{k+1}]$
  - Particle composition:
    $\vec{F_c} = (f_1, f_2, , \cdots, f_{nc-1})$
    f_s is mass fraction of component s;
    nc is total number of components;
- Aerosol Dynamic:
  - Nucleation: *binary nucleation of sulphate and water (Vehkamäki et al.,2002)*
  - Coagulation: *externally mixed coagulation model of Dergaoui et al. (2013)*
  - Condensation/Evaporation: *newly developed*



Size-composition discretization (external)



Size discretization (internal)

Model description

- Dynamic Equations for Condensation/Evaporation (C/E):

External:

$$\frac{\partial N^j}{\partial t} = 0$$

$$\frac{\partial Q_i^j}{\partial t} = \int_{m_k^-}^{m_k^+} \int_{f_{gi}^-}^{f_{gi}^+} \bar{n} \; I_i \; dm df_i \sim N^j \; I_i$$

Internal:

$$\frac{\partial N^k}{\partial t} = 0$$

$$\frac{\partial Q_i^j}{\partial t} = \int_{m_k^-}^{m_k^+} \bar{n} \; I_i \; dm \sim N^k \; I_i$$

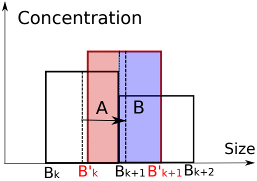
$$I_i = \frac{\partial m_i}{\partial t} = 2\pi \; D_i^g \; d_p \; f(K_n, \alpha_i)(c_i^g(t) - c_i^s(\vec{x}, t))$$

D_i^g: *molecular diffusivity in the air.*
d_p: *particle wet diameter*
c_i^g: *gas phase concentration of species* X_i
The non-continuous effects:
$f(K_n, \alpha_i)$: *Knudsen number* K_n*; accommodation coefficient* α_i

- Surface equilibrium concentration c_i^s:
Inorganic: *ISORROPIA (Nenes et al., 1998);*
Organic: $\mathbf{H^2O}$ *(Hydrophilic/Hydrophobic Organic)(Couvidat et al., 2012)*

Model description

- Size redistribution:
  HEMEN (Devilliers et al., 2013)
  or
  Moving center (Jacobson, 1997)
- Composition redistribution:
  Moving center



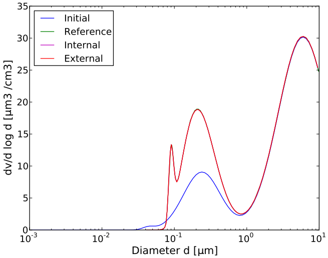
Size redistribution

- Numerical implementation:
  - Coagulation and C/E + nucleation are split
  - Solver: Second-order Rosenbrock (ROS2) (Verwer et al., 1999)

Model validation by comparison to internal mixing

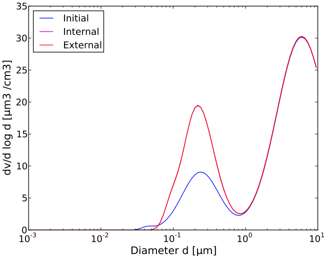
- Initial conditions:
  - Regional hazy scenario (Zhang et al., 1999)
  - Emission: *Sulphuric acid 5.5* $\mu m^3 cm^{-3}$ *per 12 hours*
  - Simulation time: *12 hours*
  - Discretization: *100 size bins, 10 composition bins*
  - Composition: 50% sulphate (non-volatile)
    50% dust with same density (1.8 $\mu g/cm^3$)

C/E Only



Volume distribution

C/E + Coagulation

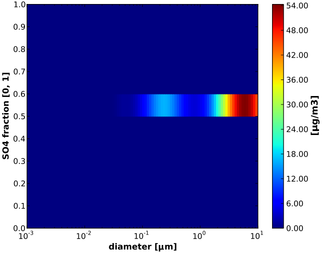


Volume distribution

Model validation by comparison to internal mixing

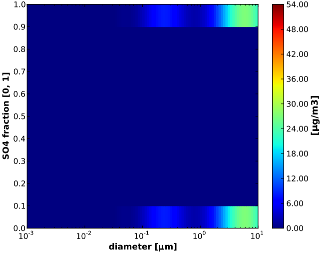
- Comparison between internal and external cases for C/E + coagulation:
  Particle mass concentration as a function of particle size and composition:

Internal mixing



Initial

External mixing

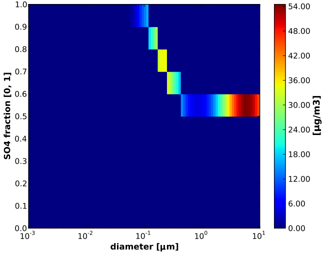


Initial

Model validation by comparison to internal mixing

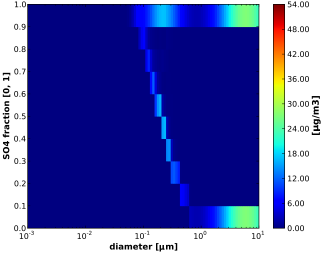
- Comparison between internal and external cases for C/E + coagulation:
  Particle mass concentration as a function of particle size and composition:

Internal mixing



After 12 hours simulation

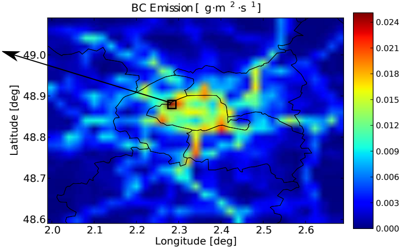
External mixing



After 12 hours simulation

0D test with realistic data

- Initial conditions:
  - Extract concentrations, emissions and meteorology at (lon, lat)=(2.28°,48.88°)
  - Emissions: *during 44 min (no dilution nor advection)*
  - Simulation time: *12 hours*
    Start: 2 AM, 01/07/2009
  - Composition: *31 species, 5 groups*
  - Discretization: *7 size bins, 3 composition fractions per group of species*
  - Size redistribution: *Moving center*



BC emission map at 2 AM (01/07/2009) over Paris

- Group identification:
  1. HydrophiLic Inorganic group (HLI): *sodium, sulphate, nitrate, ammonium, chloride*
  2. HydrophiLic Organic group (HLO): *9 hydrophilic organic species*
  3. HydrophoBic Organic group (HBO): *14 hydrophobic organic species (Couvidat et al., 2012)*
  4. Black Carbon group (BC): *black carbon*
  5. DUst group (DU): *soil, dust and fine sand*

0D test with realistic data

- Composition discretization:
  - Three composition fraction sections: [0%-20%],[20%-80%],[80%-100%]
  - 20 particle types, 5 unmixed types, 15 mixed types

20 Externally Mixed Particle Types

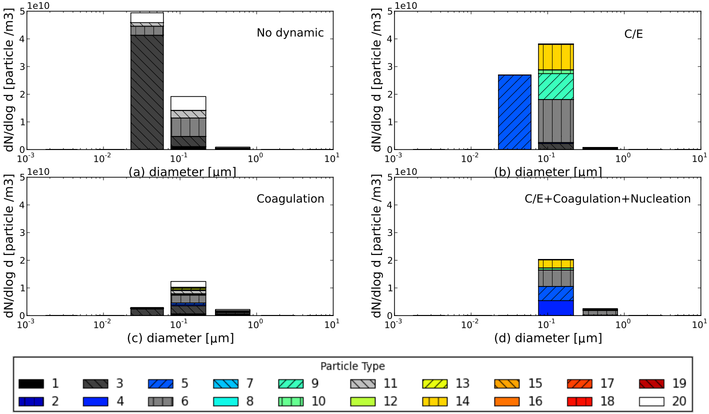
Color code	Type index	Mixing state	Mass fraction of each groups (%)				
			HLI	HLO	HBO	BC	DU
	1	Unmixed (DU)	0-20	0-20	0-20	0-20	0-100
	2	Mixed	0-20	0-20	0-20	20-80	0-100
	3	Umixed (BC)	0-20	0-20	0-20	80-100	0-100
	4	Mixed	0-20	0-20	20-80	0-20	0-100
	5	Mixed	0-20	0-20	20-80	20-80	0-100
	6	Unmixed (HBO)	0-20	0-20	80-100	0-20	0-100
	7	Mixed	0-20	20-80	0-20	0-20	0-100
	8	Mixed	0-20	20-80	0-20	20-80	0-100
	9	Mixed	0-20	20-80	20-80	0-20	0-100
	10	Mixed	0-20	20-80	20-80	20-80	0-100
	11	Unmixed (HLO)	0-20	80-100	0-20	0-20	0-100
	12	Mixed	20-80	0-20	0-20	0-20	0-100
	13	Mixed	20-80	0-20	0-20	20-80	0-100
	14	Mixed	20-80	0-20	20-80	0-20	0-100
	15	Mixed	20-80	0-20	20-80	20-80	0-100
	16	Mixed	20-80	20-80	0-20	0-20	0-100
	17	Mixed	20-80	20-80	0-20	20-80	0-100
	18	Mixed	20-80	20-80	20-80	0-20	0-100
	19	Mixed	20-80	20-80	20-80	20-80	0-100
	20	Unmixed (HLI)	80-100	0-20	0-20	0-20	0-100

Results for number concentration:

Emissions: Organics (gray), Dust (black), Inorganics (white)

Mixing state after simulation based on number concentration

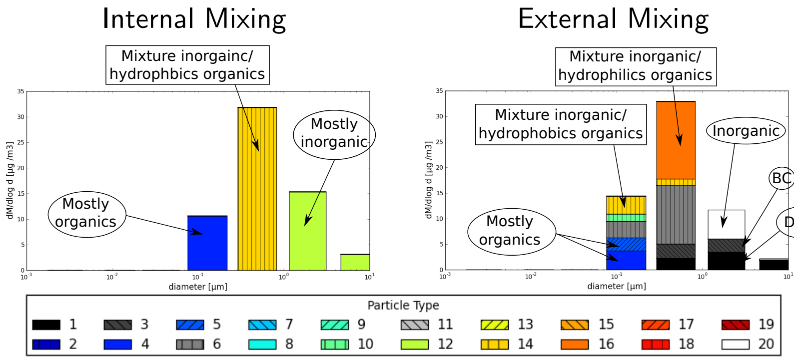
Process	No Dynamic	C/E	Coagulation	C/E+Coag+Nucl
Praticle mixed (%)	0	72	22	66



Condensation is more efficient at mixing particles than coagulation

0D test with realistic data

- Total mass: Internal: $35.76 \mu g/m^3$; External: $35.75\ \mu g/m^3$;
- Particle composition differs whether internal or external mixing
- White, gray and black: particles are not mixed
  - White = inorganic
  - Gray = organics
  - Black = dust



Particle mass distribution after simulation

Conclusions and future work

- Conclusions:
  - SCRAM simulates the aerosol dynamics of externally mixed particles (Condensation/evaporation, coagulation and nucleation)
  - Validation by comparison to internal mixing simulations
  - External and internal mixing lead to different particle compositions and size distribution
- Future Work:
  - Integration into the air quality modeling platform POLYPHEMUS for 3D simulation. Investigate its performance in modeling air quality by comparing to observations of the MEGAPOLI[1] campaign.

[1] Megacities: Emissions, urban, regional and Global Atmospheric POLlution and climate effects, and Integrated tools for assessment and mitigation