Marine boundary layer aerosol in Eastern North Atlantic: seasonal variations and the key controlling processes

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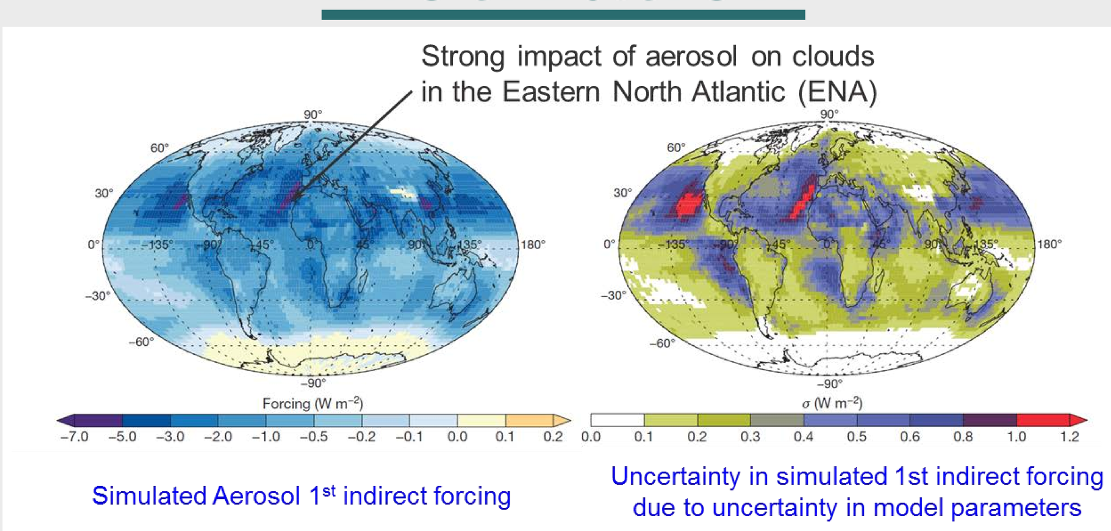
Motivation



Adapted from Carslaw et al. (2013)

- Marine low clouds are important in climate predictions, and are particularly susceptible to perturbations in aerosols
- It is imperative to understand marine boundary layer (MBL) aerosol properties both under natural condition and the changes associated with continental emissions.
- ENA has strong but poorly constrained aerosol-cloud interactions and diverse but persistent marine low clouds

Measurements

Three-year (2015 ~2017) data collected at ENA site by Atmospheric Radiation Measurement (ARM) Climate Research

Properties Measured	Symbol	Unit	Instruments	Time resolution	Measurement period
Total aerosol number concentration	CN	cm^{-3}	CPC 3772 (TSI)	1s	Oct. 2013 to Aug. 2014, June 2015 to present
Aerosol number size distribution in 55 nm to 1 μm	$dN/d\ln D_p$	cm^{-3}	UHSAS-0.055 (DMT)	10s	Feb. 2014 to present
CCN number at five supersaturations (ss)[a]	CCN (ss%)	cm^{-3}	CCN-100 (DMT)	1s, ss level changes every ~12 min	Oct. 2013 to Apr. 2015, July 2016 to present
Aerosol Absorbing coefficient	B_{abs}	Mm^{-1}	3λ PSAP (Radiance Research)	1s, inlet changes between 1 μm and 10 μm every 30 min	Oct. 2013 to present
Aerosol Scattering coefficient	B_{sca}	Mm^{-1}	Nephelometer 3563 (TSI)		Jan. 2014 to present
trace gases of CO, NO_2 and H_2O	/	ppb	Gas Analyzer 48C (Thermo Electron)	1s	April 2015 to present
trace gas of O_3	/	ppb	Ozone monitor 49i (Thermo Fisher)	1s	Oct. 2013 to present
Meteorology parameters[b]	/	/	ENA Aerosol Observing System	1s	Jan. 2014 to present
Precipitation rate at cloud base	P_{CB}	$mm\ h^{-1}$	Retrived	30min	
MBL height	H_{MBL}	m	Ceilometer CL31 (Vaisala)	16s	Dec. 2014 to present
Cloud thickness	h	m			
Cloudy time fraction	t_{cloud}	1			

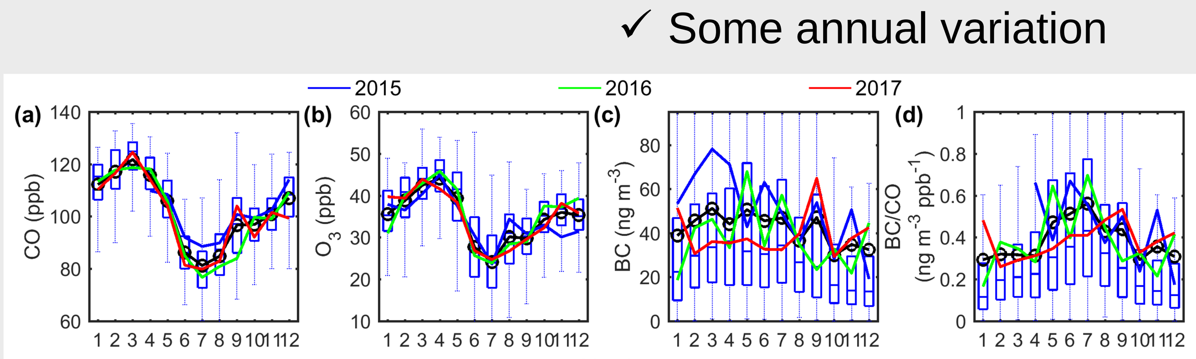
Results

Trace Gas (CO & O_3)

- ✓ Indicator of entrainment from free troposphere (FT) at ENA
- ✓ Low in summer, high in spring-winter
- ✓ No annual variation

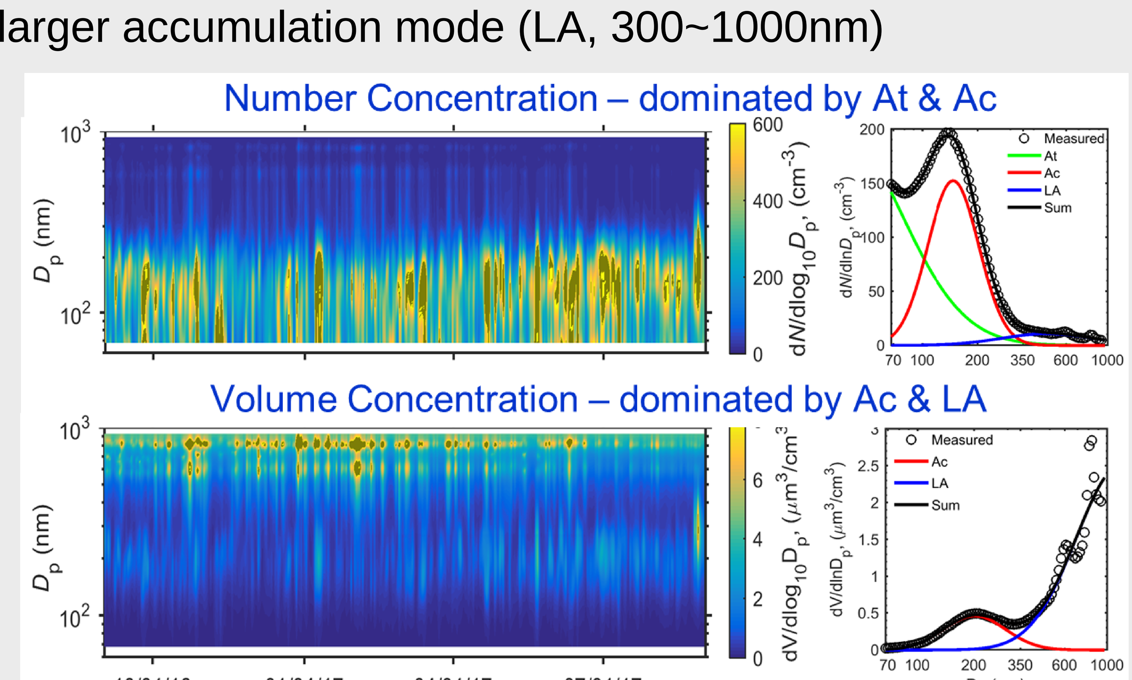
Absorbing Aerosol (BC)

- ✓ Indicator of overall effect of FT and precipitation scavenging (PRCP)
- ✓ Relatively weak seasonal variations
- ✓ Some annual variation

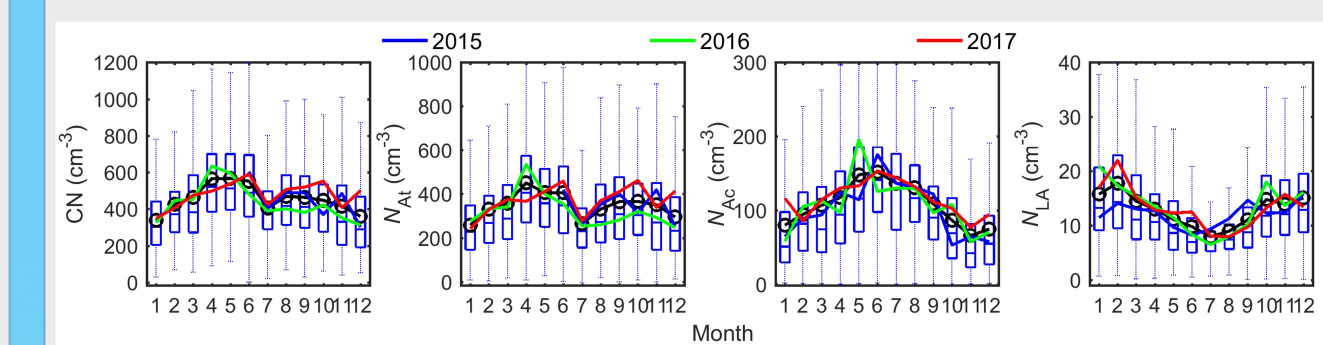


Modes of Aerosol Size Distribution

- ✓ Aitken mode (At, <100 nm)
  - not well constrained; N_{At} = CN- N_{Ac} – N_{LA}
- ✓ Accumulation mode (Ac, 100~300 nm)
- ✓ larger accumulation mode (LA, 300~1000nm)



Seasonal Variation of Aerosol Size Distribution



CN and N_{At}
- ✓ weak seasonal variation
- ✓ some annual variation

N_{Ac}
- ✓ Summer high, winter low
- ✓ Annual stable

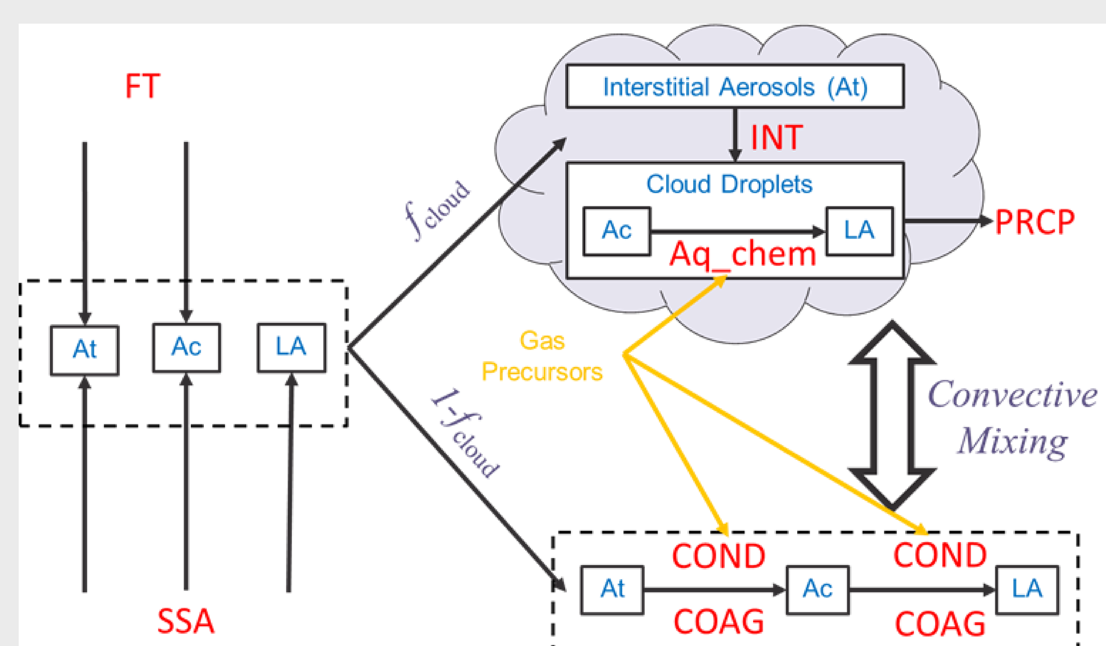
N_{LA}
- ✓ Summer low, winter high
- ✓ Annual stable

Governing Equation of MBL Aerosol

$$\frac{\partial N}{\partial t} = \frac{\partial N}{\partial t}\bigg|_{FT} + \frac{\partial N}{\partial t}\bigg|_{SSA} + \frac{\partial N}{\partial t}\bigg|_{NCP} + \frac{\partial N}{\partial t}\bigg|_{CP}$$

$$\frac{\partial N}{\partial t} = \frac{\partial N}{\partial t}\bigg|_{FT} + \frac{\partial N}{\partial t}\bigg|_{SSA} + \frac{\partial N}{\partial t}\bigg|_{COND} + \frac{\partial N}{\partial t}\bigg|_{COAG} + \frac{\partial N}{\partial t}\bigg|_{AQ\_CHEM} + \frac{\partial N}{\partial t}\bigg|_{PRCP} \quad \text{,for CCN}$$

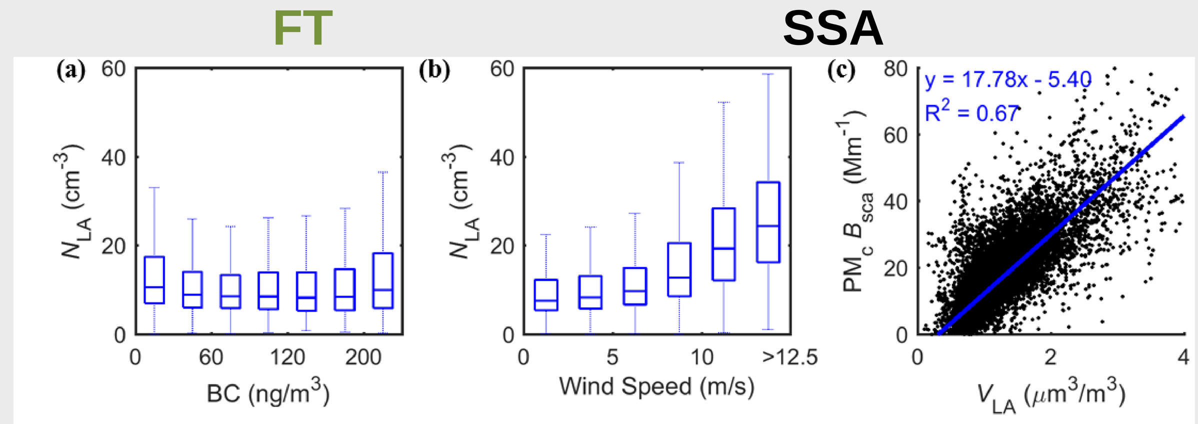
$$\frac{\partial N}{\partial t} = \frac{\partial N}{\partial t}\bigg|_{FT} + \frac{\partial N}{\partial t}\bigg|_{SSA} + \frac{\partial N}{\partial t}\bigg|_{COND} + \frac{\partial N}{\partial t}\bigg|_{COAG} + \frac{\partial N}{\partial t}\bigg|_{INT} \quad \text{,for non-CCN}$$



Key Processes of LA mode

$$\frac{\partial N_{LA}}{\partial t} = \frac{\partial N_{LA}}{\partial t}\bigg|_{FT} + \frac{\partial N_{LA}}{\partial t}\bigg|_{SSA} + \frac{\partial N_{LA}}{\partial t}\bigg|_{COND} + \frac{\partial N_{LA}}{\partial t}\bigg|_{COAG} + \frac{\partial N_{LA}}{\partial t}\bigg|_{AQ\_CHEM} + \frac{\partial N_{LA}}{\partial t}\bigg|_{PRCP}$$

FT **SSA**



N_{LA} ~ BC: poor
- ✓ Less likely dominated by continental emissions in FT

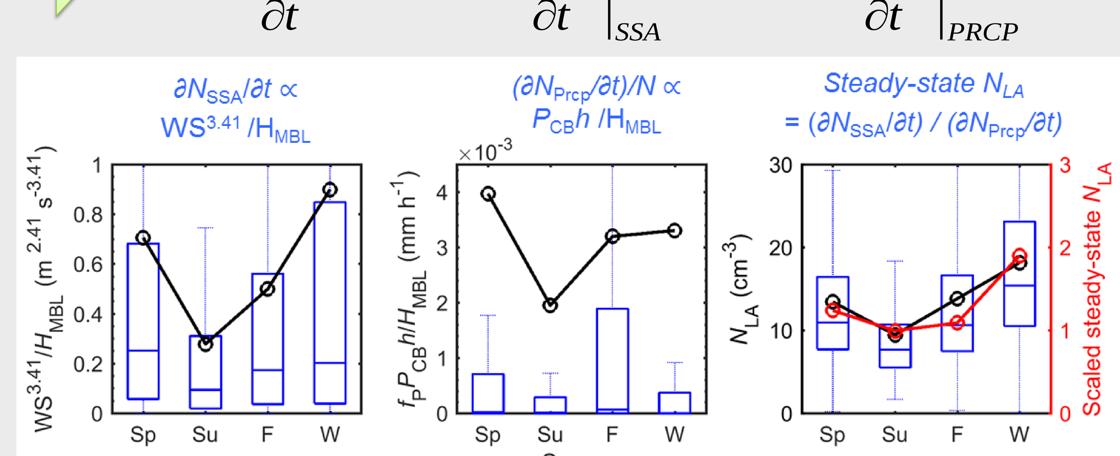
N_{LA} ~ WS: positive
- ✓ SSA production flux F ∝ $WS^{3.41}$

V_{LA} ∝ PM_c: strong
- ✓ common source
- ✓ PMc dominated by SSA

Condensation, Coagulation and cloud chemistry

- ✓ Not important by calculation

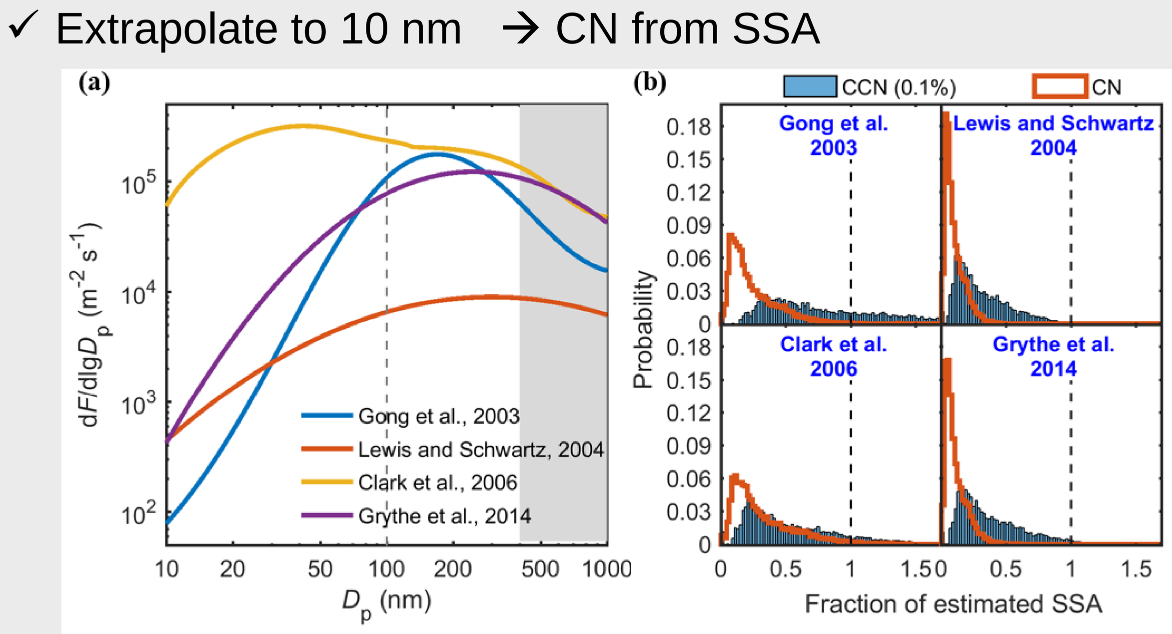
$$\frac{\partial N_{LA}}{\partial t} = \frac{\partial N_{LA}}{\partial t}\bigg|_{SSA} + \frac{\partial N_{LA}}{\partial t}\bigg|_{PRCP}$$



Key Processes of At and Ac modes

Minor SSA contribution to At – Ac mode

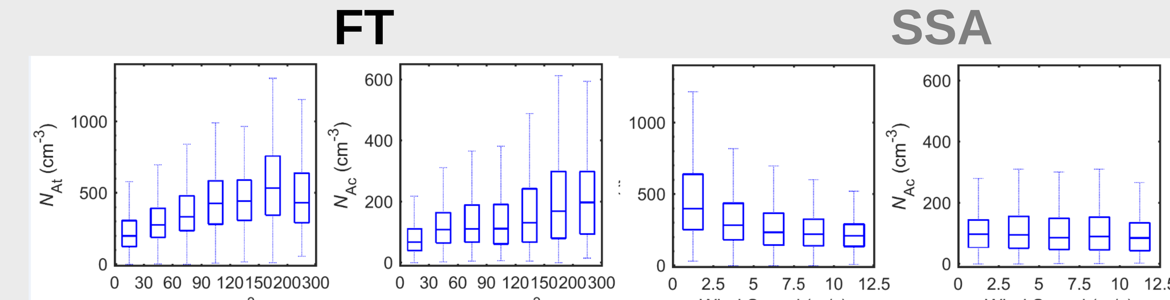
- SSA estimation by production flux curve and N_{LA}
- ✓ Assume N with Dp > 400nm are all SSA and a conserved shape of SSA production flux
- ✓ Extrapolate to 100 nm → CCN(0.1%) from SSA
- ✓ Extrapolate to 10 nm → CN from SSA



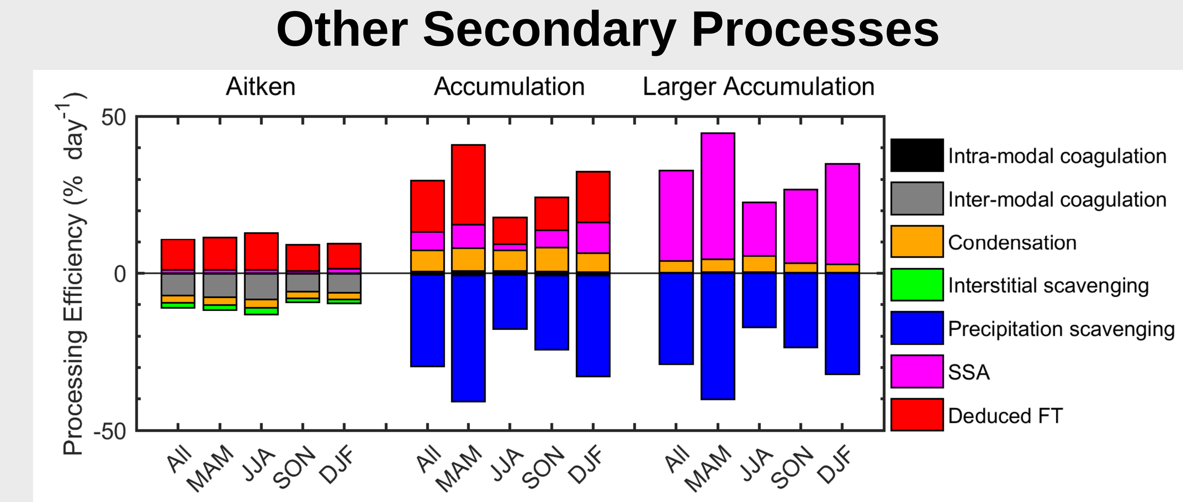
Minor annual contribution of SSA to N_{Ac} (20.6 %) and N_{At} (9.9 %)

Key Processes of At and Ac mode

FT **SSA**



Other Secondary Processes



$$\frac{\partial N_{Ac}}{\partial t} = \frac{\partial N_{Ac}}{\partial t}\bigg|_{FT} + \frac{\partial N_{Ac}}{\partial t}\bigg|_{SSA} + \frac{\partial N_{Ac}}{\partial t}\bigg|_{COND} + \frac{\partial N_{Ac}}{\partial t}\bigg|_{PRCP}$$

$$\frac{\partial N_{At}}{\partial t} = \frac{\partial N_{At}}{\partial t}\bigg|_{FT} + \frac{\partial N_{At}}{\partial t}\bigg|_{COND} + \frac{\partial N_{At}}{\partial t}\bigg|_{COAG} + \frac{\partial N_{At}}{\partial t}\bigg|_{INT}$$

Summary

- Aerosols at ENA typically consist of 3 modes: At, Ac and LA, which showed different seasonal variations
- N_{LA} is dominated by SSA and precipitation scavenging, the overall effect being higher in winter and lower in summer
- SSA often contributed a minor fraction of N_{At} (annual average 9.9 %) and N_{Ac} (annual average 20.6 %)
- Controlling processes of N_{Ac} and N_{At} are more complex. Sources and sinks for At and Ac modes showed similar seasonal pattern, resulting in their weak seasonal variation