Arctic fog chemistry induces the unexpected growth of Aitken mode particles to CCN-active particles

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Introduction and Motivation

New particle formation (NPF) and early growth are efficient processes producing high concentrations of cloud condensation nuclei (CCNs) precursors in the Arctic marine boundary layer (AMBL) (1). However, due to short lifetime and lack of condensable vapors, newly formed particles do often not grow beyond 50 nm causing low CCN particle concentrations in the AMBL. Thus, even the smallest Aitken mode particle growth is capable to significantly increase the CCN budget. The emission of trace gases from the Arctic ocean into the atmosphere and their further oxidation is crucial to understand the formation and further growth of particles.
During the PASCAL campaign (May to July 2017), aerosol particles measurements were performed on the research vessel Polarstern (2) and an unexpected rapid growth of Aitken mode particles was observed after fog episodes. Combined field data analyses and detailed multiphase chemistry box model simulations with CAPRAM were performed to study the underlying processes and to investigate if this growth is related to chemical processes within the Arctic fog.

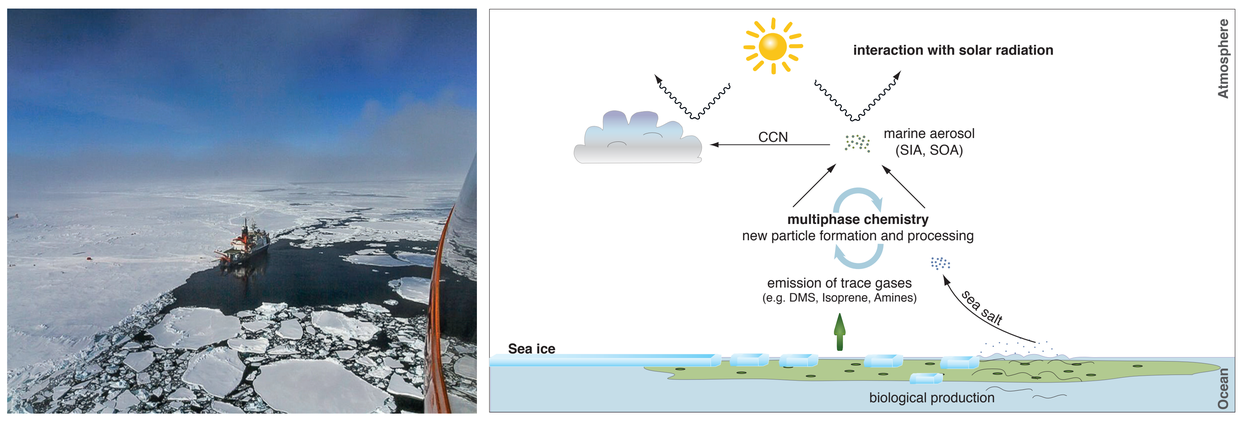

Fig. 1: Overflight over the R/V Polarstern during PASCAL (left, photo: Stephan Schön, Sächsische Zeitung). Scheme of the importance of trace gases and multiphase chemistry for Arctic particle & CCN concentration, and climate (right).

Measurement of unexpected rapid growth event

- Rapid unexpected growth of Aitken mode particles observed after fog between 1st to 2nd and 19th to 20th of June during PASCAL campaign
- After fog dissipation, rapid growth of Aitken mode particles with > 20 nm h^{-1} into Accumulation mode observed
- Growth could only be observed after sequence of:

  (i) New particle formation and

  (ii) fog formation, long fog processing of the air parcel, and dissipation

Fig. 2: (A) Particle number and (B) surface area size distribution, as well as integrated (in a size range from 10 to 800 nm) particle number, and surface area concentrations for Aitken (10 to 60 nm; solid line) and accumulation (60 to 800 nm; dashed line) mode particles. Fog episode duration (visibility < 1000 m) is marked by the shaded area, while observed post-fog particle growth is shown as an evolution of particle geometric mean diameter (by fitting PNSD with log-normal distribution; black squares).

Multiphase chemistry model setup

- Box model studies with the air parcel model SPACCIM (SPectral Aerosol Cloud Chemistry Interaction Model; 3), for the growth event (between 1st to 2nd of June)
- Coupling gas-phase mechanism MCMv3.2, aqueous-phase mechanism CAPRAM4.0 (4), DMS chemistry module CAPRAM–DM1.0 (5) and halogen chemistry module CAPRAM–HM2.1 (6)
- Three simulations: **(i)** without fog processing, **(ii)** with fog processing, and **(iii)** with fog processing and high supersaturation (SuperSat = 0.5%)

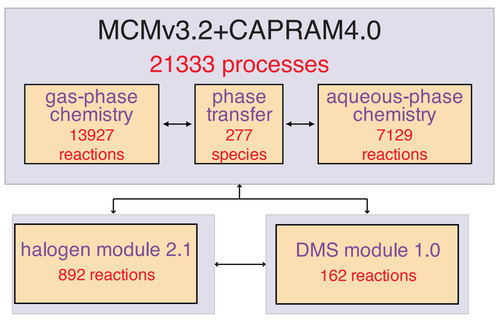

Fig. 3: Scheme of the applied multiphase chemistry mechanism.

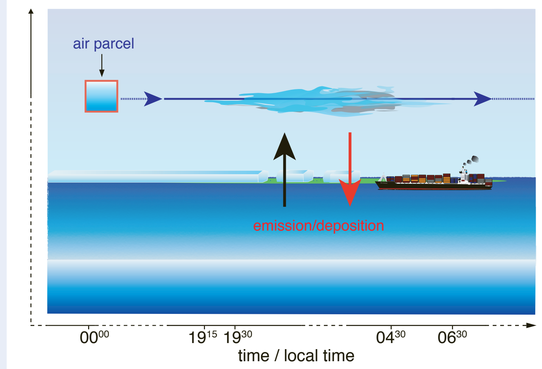

Fig. 4: Meteorological model scenario.

Model results

Effect of fog related chemistry on particle mass and composition

- Differentiation of particles into CCN-inactive, partly CCN-active and CCN-active
- CCN-active particles between > 31 to ≤90 nm diameter in simulation (ii) and ≤ 31 nm diameter in simulation (iii)
- Normalized logarithmic increase of CCN-inactive particles mass right after fog evaportation for all simulations with fog

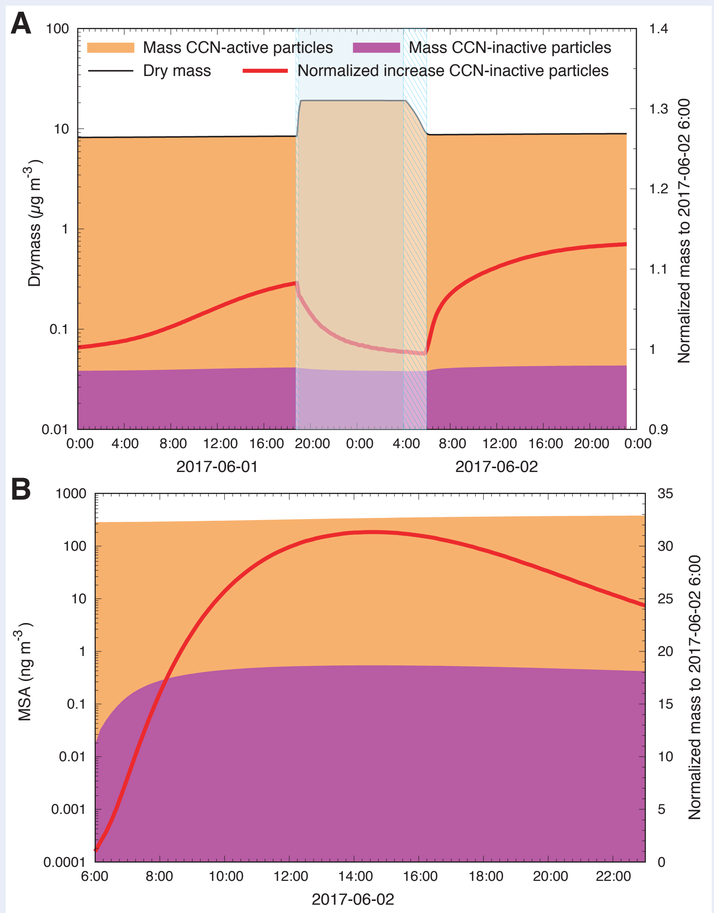

Fig. 5: (A) Modeled dry aerosol particle mass in the simulation (ii) for CCN-active and -inactive particles. The light blue bar represents the fog. The shaded more transparent blue bars show the formation and evaporation of the fog, respectively.
(B) Modeled MSA particle mass in the simulation with fog occurrence without high SuperSat for CCN-active and -inactive particles right after fog dissipation.

- CCN-inactive particles only in simulation (iii), but deactivate after about 3.7 hours
- Deactivation able to explain observed increase of particles between 10-60 nm diameter
- Long fog processing and thus possible advection of non-fog processed particles in the range of the partly CCN-active particles
- Such particles do not activate again and thus no cloud-related mass production possibl
- Investigation of concentration evolution for main compounds in marine environment
- Logarithmic increase after fog dissipation for MSA (methane sulfonic acid), ammonia and nitrate in Aitken mode particles, but not for sulfate
- Strongest change for MSA
- MSA, also nitrate and oxalic acid found to be chemically formed in fog droplets

Comparison of modeled with measured impactor stage mass

- Comparison with measurements only possible for not activated particles (IS 1)
- Very good agreement between modeled and measured MSA and nitrate for simulation (ii), but large overestimation for simulation (iii)

Table 1: Comparison between the modeled and measured MSA and nitrate in the 1st impactor stage.

	MSA / ng m^{-3}	Nitrate / ng m^{-3}
Simulation (ii)	1.41	2.08
Simulation (iii)	32.05	11.35
Measurement	1.14	bdl

Summary and Outlook

- During PASCAL campaign, an unexpected rapid growth (> 20 nm h^{-1}) of Aitken mode particles into CCN size measured after frequency of new particle formation and fog
- Combined field measurements and box model studies revealed importance of fog chemistry for mass production and growth
- Supersaturation critical for particle activation, in-fog mass production and distribution
- Multiphase chemistry produces semi-volatile compounds, particularly MSA, that evaporate after dissipation into gas phase and recondense on Aitken mode particles

References

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