

Activation and hygroscopic growth  
behavior of soot and dust coated with SOA  
or  $\text{H}_2\text{SO}_4$ : Results from IN 11 and ACI 02  
campaigns, DMT-CCNC & LACIS-mobile  
measurements

presented by  
Katrin Mildenerger  
Institute for Tropospheric Research  
in Leipzig, Germany

# Overview

- ☛ Our part in VI-ACI
- ☛ AIDA Studies
- ☛ Instrumentation
- ☛ IN 11
  - Experimental Setup
  - Results and Discussion
  - Status
- ☛ ACI 02
  - Experimental Setup
  - Methods
  - Results and Discussion
  - Summary
  - Outlook

# Our part in VI-ACI

- ☛ Characterize the cloud condensation properties of soot and mineral dust particles
- ☛ Activation and hygroscopic growth measurements of coated and uncoated soot and mineral dust particles
- ☛ Closure study between hygroscopic growth and activation properties
- ☛ Studying the effect of the coating of particles on their hygroscopic growth and activation behavior

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

-Outlook

# AIDA Studies

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

-Outlook

IN 11 (measurements and analysis, Ziese et al.)

☛ Investigated aerosol types:

- Uncoated GFG-soot (nitrogen or argon as carrier gas)
- GFG-soot coated with succinic acid
- CAST-soot with different organic carbon contents uncoated and coated with sulfuric acid or succinic acid

ACI 02 (measurements and analysis, Mildenberger et al.)

☛ Investigated aerosol types:

- Uncoated mineral particles (glass spheres, BCR and ATD)
- Coated mineral particles with SOA or sulfuric acid
- GFG-soot coated with SOA

# Instrumentation

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion  
-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

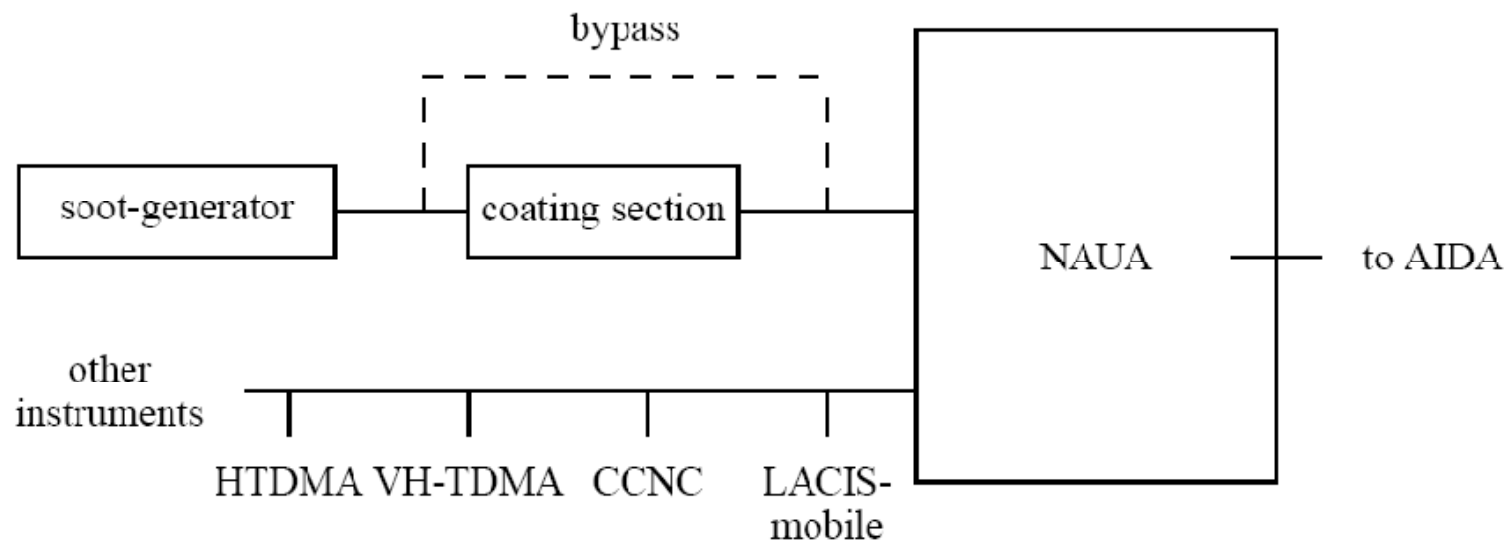
-Outlook

- ☛ [HTDMA, hygroscopic growth; FZ Jülich, Germany]
- ☛ [VH-TDMA, volatile and hygroscopic behavior; Aubire, France]
- ☛ LACIS-mobile (hygroscopic growth up to 99.39 % RH)
- ☛ Cloud Condensation Nucleus Counter (activation measurements 0.07 – 1.6 % SS)



# IN 11 - Experimental Setup

- All results for monodisperse aerosol sampled from the NAUA-chamber



Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

-Outlook

# IN 11 – Results and Discussion

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

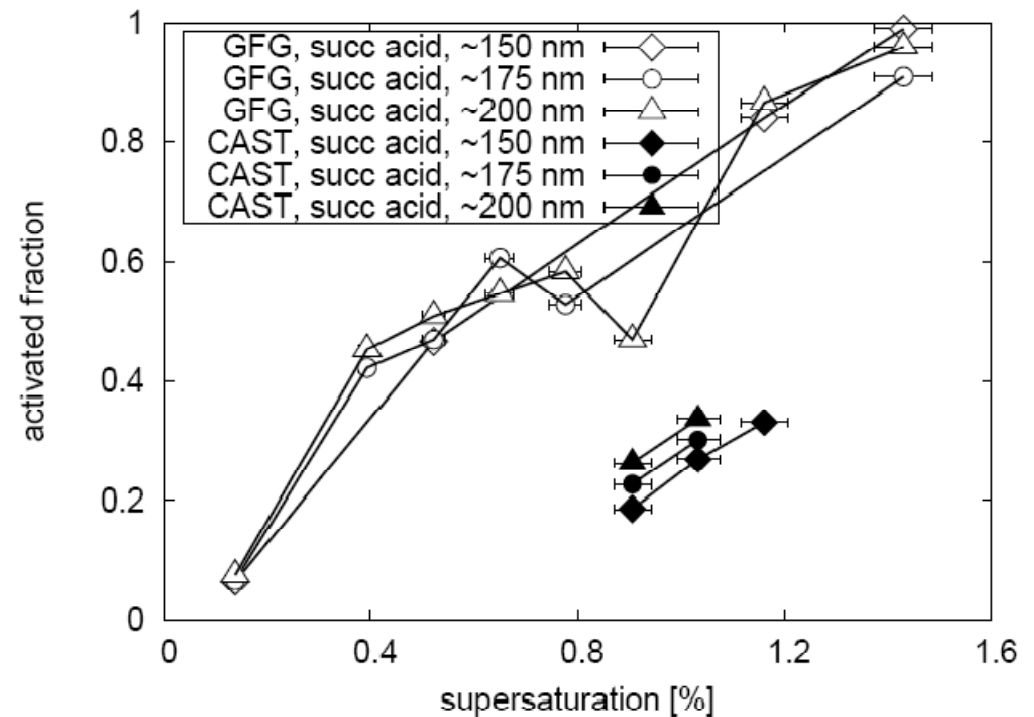
-Methods

-Results and  
Discussion

-Summary

-Outlook

- No hygroscopic growth for all pure soot types
- No activation up to 1.6% except GFG with Ar as carrier gas
- No hygroscopic growth for all soot types coated with succinic acid (after wetting shrinking of the particles was observed)
- Activation of the different soot types coated with succinic acid was observed (see Figure)
- Only 54% of the particles coated (non uniform coating)???



# IN 11 – Results and Discussion

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

-Outlook

- Hygroscopic growth for CAST-soot coated with sulfuric acid was found  
→ Hygroscopic growth increased with increasing OC content (OC content included PAHs (cf. AMS data))

Hypothesis: insoluble PAHs reacted with sulfuric acid to smaller and more soluble molecules

→ VHTDMA, HTDMA, LACIS showed slightly different results

Hypothesis: different coating thicknesses for different dry particle sizes as the coated particles were polydisperse

- Activation of CAST-soot coated with sulfuric acid was possible  
→ no change of the activation behavior with increasing OC content



# IN 11 – Status

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

-Outlook

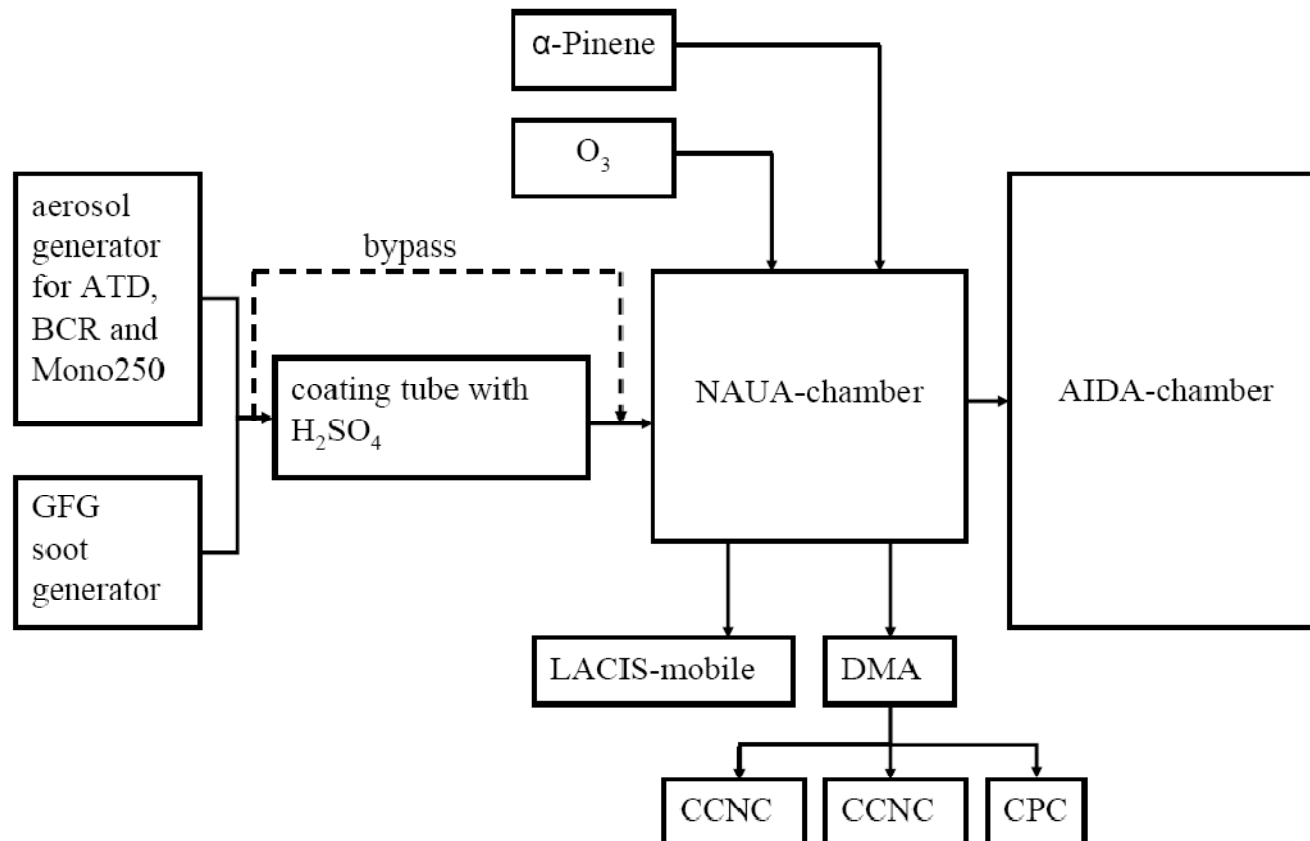
Paper is in preparation:

“Hygroscopic growth and activation of soot particles: uncoated, succinic or sulfuric acid coated”

Ziese et al., 2010

# ACI 02 - Experimental Setup

- All results for monodisperse aerosol sampled from the NAUA-chamber



# ACI 02 – Methods

- ☛ Determination of the critical supersaturation ( $S_{crit}$ ) from CCNC measurements
- ☛ Calculation of the coating thickness: Köhler model with insoluble core and one respectively two soluble compounds was used assuming spherical particles ( $T = 20^{\circ}\text{C}$ ,  $\sigma$  of water)
- ☛ Closure study: Prediction of critical supersaturation using values of the hygroscopicity parameter  $\kappa$  derived from hygroscopic growth measurements

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

-Outlook

# ACI 02 – Results and Discussion

(Activation of uncoated and coated Mono250)

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

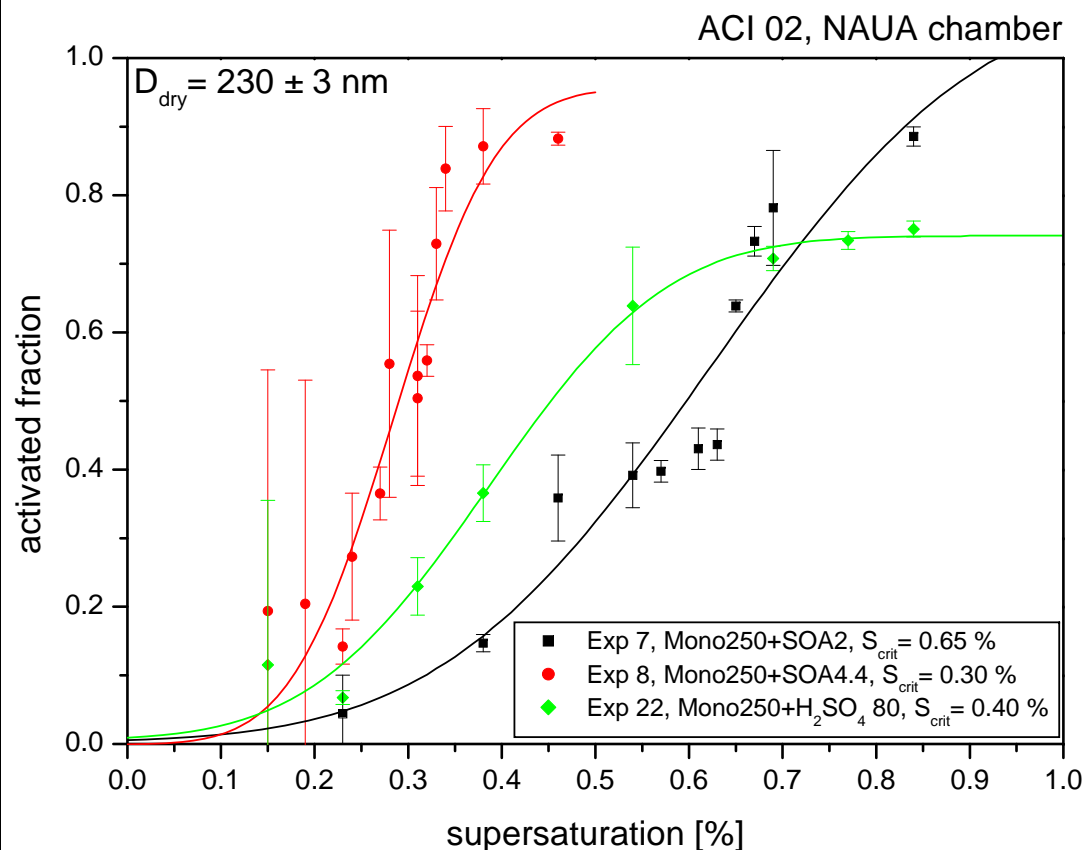
-Methods

-Results and  
Discussion

-Summary

-Outlook

- No activation of pure glass spheres (Mono250) for supersaturations up to 1.31%
- After coating activation was observed



- The larger the coating thickness, the easier the particles activated
- Hygroscopicity ( $\kappa$ ) increased with coating thickness
- Coating thickness:
  - Exp 7  $\rightarrow$  0.3 nm
  - Exp 8  $\rightarrow$  4.5 nm
  - Exp 22  $\rightarrow$  0.2 nm

# ACI 02 – Results and Discussion

(Activation of uncoated and coated BCR)

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

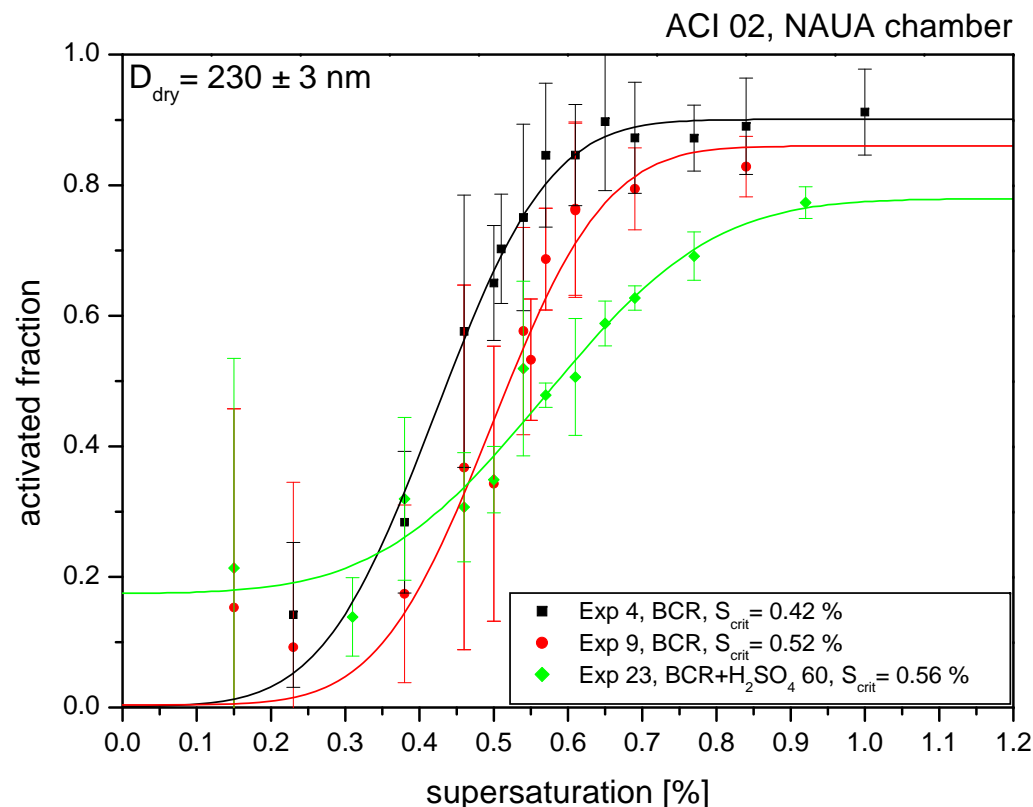
-Methods

-Results and  
Discussion

-Summary

-Outlook

Activation of pure BCR particles was observed



Critical supersaturation needed for activation was higher for particles coated with sulfuric acid than for pure BCR particles  
Explanation???

# ACI 02 – Results and Discussion

(Activation of uncoated and coated ATD)

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion  
-Status

ACI 02

-Experimental  
Setup

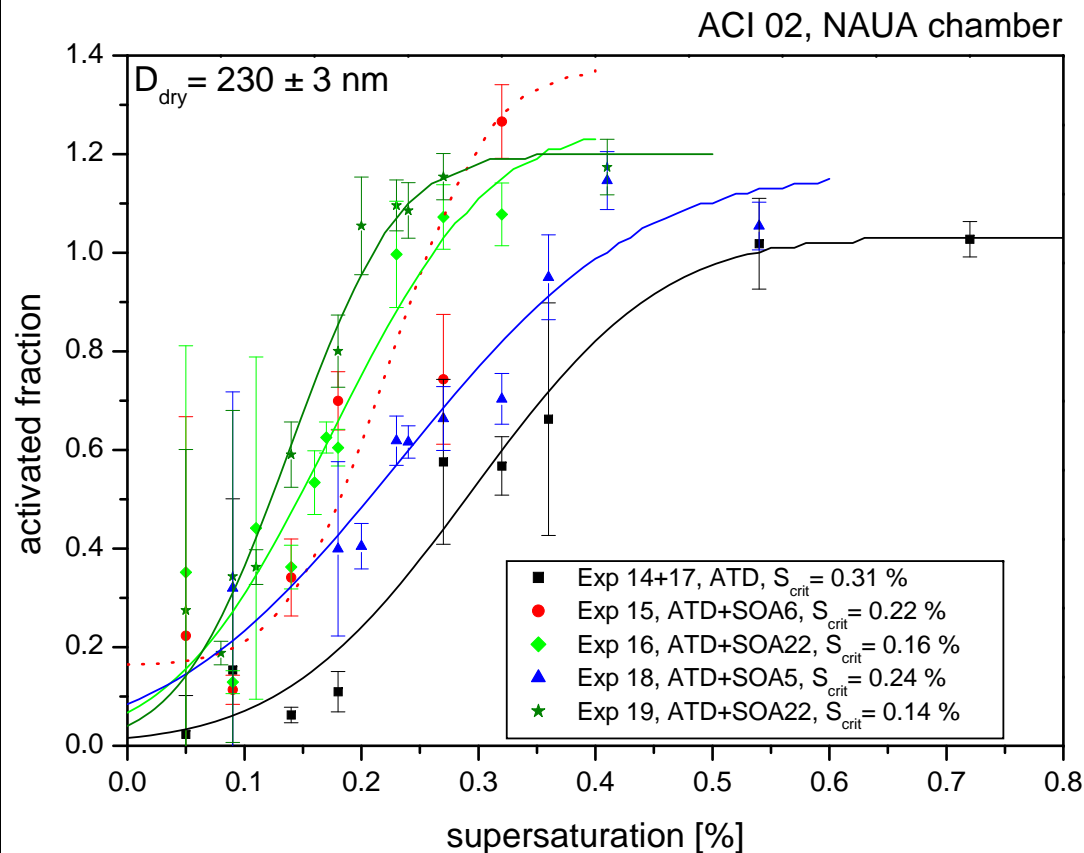
-Methods

-Results and  
Discussion

-Summary

-Outlook

Activation of pure ATD particles was observed



The larger the coating thickness, the easier the particles activated

Hygroscopicity ( $\kappa$ ) increased with coating thickness

Coating thickness:

- Exp 15  $\rightarrow$  8.9 nm
- Exp 16  $\rightarrow$  18.4 nm
- Exp 18  $\rightarrow$  7.2 nm
- Exp 19  $\rightarrow$  27.7 nm

# ACI 02 – Results and Discussion

(Activation of uncoated and coated ATD)

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

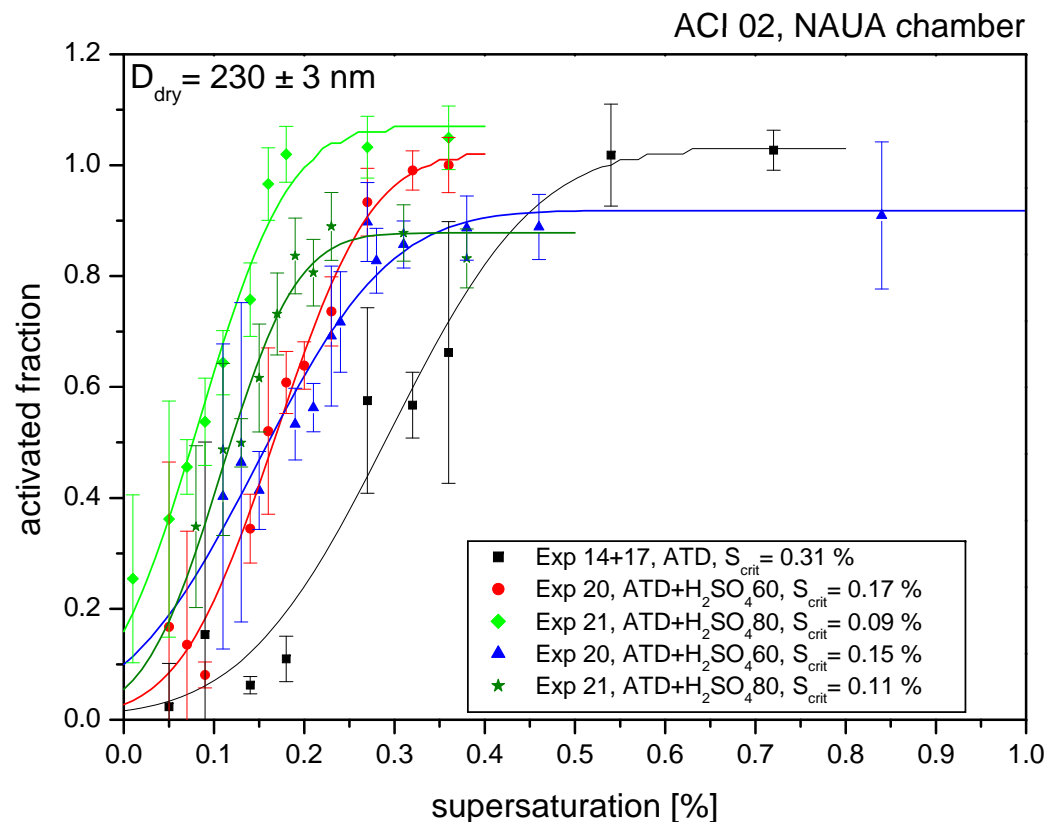
-Methods

-Results and  
Discussion

-Summary

-Outlook

Activation of pure ATD particles was observed



The larger the coating thickness, the easier the particles activated

Hygroscopicity ( $\kappa$ ) increased with coating thickness

Coating thickness:

- Exp 20<sub>CCNC1</sub> → 2 nm
- Exp 21<sub>CCNC1</sub> → 4 nm
- Exp 20<sub>CCNC2</sub> → 1.5 nm
- Exp 21<sub>CCNC2</sub> → 6 nm

# ACI 02 – Results and Discussion

(Activation of uncoated and coated GFG)

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

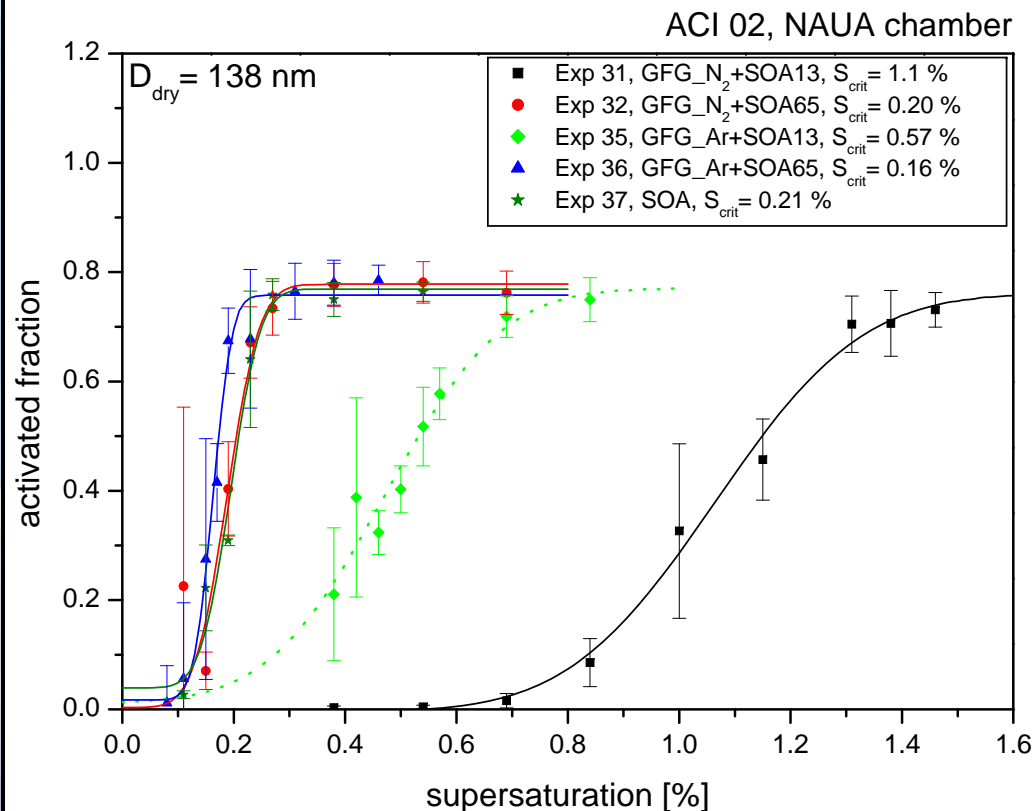
-Methods

-Results and  
Discussion

-Summary

-Outlook

- No activation of pure soot particle (GFG) and soot particles coated with a thin layer of SOA for supersaturations up to 1.31%



- The larger the coating thickness, the easier the particles activated; In exp 32 & 36 particles activated easier than pure SOA???

- Hygroscopicity ( $\kappa$ ) increased with increasing coating thickness

- Coating thickness:

- Exp 31 → 0.3 nm
- Exp 32 → < pure SOA
- Exp 35 → 3.5 nm
- Exp 36 → < pure SOA



# ACI 02 – Results and Discussion

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

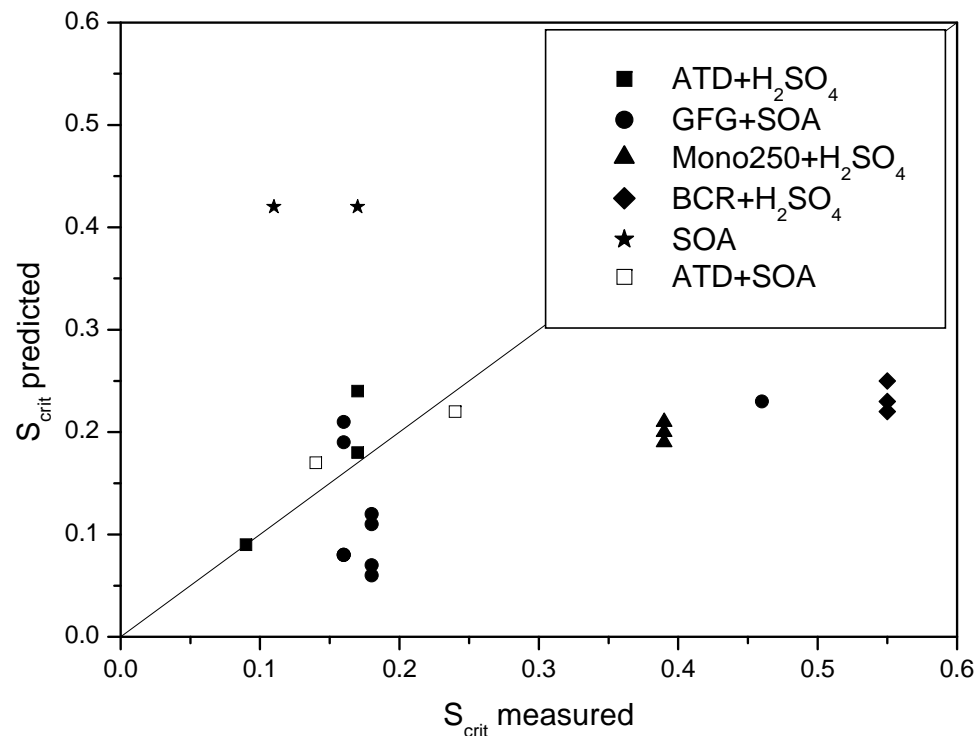
-Methods

-Results and  
Discussion

-Summary

-Outlook

- ☛ Closure study: prediction of  $S_{crit}$  from hygroscopic growth measurements by using  $\kappa$ -Köhler theory



- ☛ BCR and Mono250 particles coated with H<sub>2</sub>SO<sub>4</sub>:  
Measured  $S_{crit}$  much higher than the predicted ones
- ☛ Pure SOA:  
Measured  $S_{crit}$  much lower than the predicted ones
- ☛ ATD coated with SOA or H<sub>2</sub>SO<sub>4</sub>:  
Measured and predicted  $S_{crit}$  in good agreement
- ☛ GFG particles coated with SOA → no clear trend

# ACI 02 – Results and Discussion

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup  
-Results and  
Discussion  
-Status

ACI 02

-Experimental  
Setup  
-Methods  
-Results and  
Discussion  
-Summary  
-Outlook

Further results (not shown):

- ☛ Comparison of both CCNC → good agreement
- ☛ Additionally, for ATD the influence of dynamic shape factor (1.5) on the results was checked → absolute  $\kappa$  values and coating thicknesses differed, but trends in hygroscopic growth and activation behavior remained
- ☛ For all SOA coated particles critical supersaturation ( $S_{crit}$ ) and surface tension ( $\sigma$ ) at the activation point was iterated following Ziese et al. (2008) based on LACIS and CCNC data →  $0.026 < \sigma_{activation} < 0.060 \text{ N m}^{-1}$  (too low)

# ACI 02 – Summary

- ☛ Soluble coating of an insoluble particle generally made the particles more hygroscopic  
Except: The activation of pure BCR particles was better than the activation of BCR particles coated with sulfuric acid
- ☛ For GFG particles coated with a high amount of SOA the activation was better than for pure SOA particles
- ☛ Closure between hygroscopic growth measurements was only possible for ATD particles

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

-Outlook

# ACI 02 – Outlook

Our part in  
VI-ACI

AIDA Studies

Instrumentation

IN 11

-Experimental  
Setup

-Results and  
Discussion

-Status

ACI 02

-Experimental  
Setup

-Methods

-Results and  
Discussion

-Summary

-Outlook

## Further plans:

- More detailed consideration of the closure study results
- Implementation of a dynamic shape factor for BCR and GFG particles into calculation of  $\kappa$  and coating thicknesses
- Further discussion of the results
- Preparation of paper

## Further information needed:

- measured dynamic shape factor
- determined coating thickness from AMS measurements
- number size distributions of the aerosol particles in the NAUA-chamber

Thanks to all participants!

