The observed impact of aerosols on cloud droplet formation during the RACLETS campaign

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European Research Council
Established by the European Commission

EGU 2020
Session **AS1.24** (Clouds, aerosols, radiation and precipitation)

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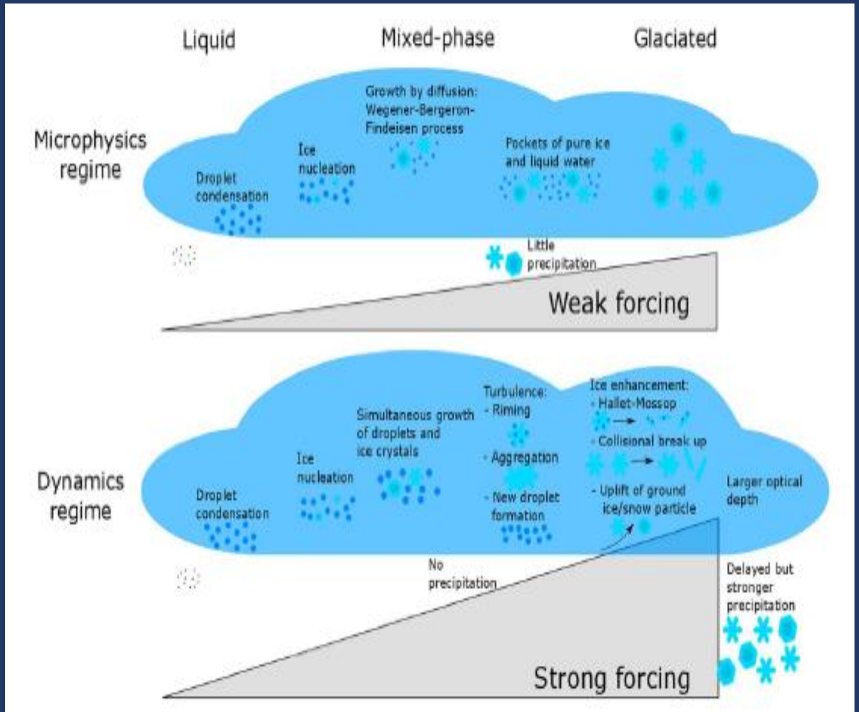
Motivation



Source: Lohmann et al. 2016

How important the aerosol concentration can be for Alpine orographic Mixed-Phase Clouds (MPCs) ?

- Aerosols and their effects on cloud microphysical properties play a key role in the formation and distribution of precipitation over **complex terrain**.
- The effect of aerosol particles on orographic precipitation remains **uncertain** due to many possible cloud **microphysical pathways**, which the hydrometeors can undergo in MPCs.
- Alpine MPCs are strongly affected by **dynamics**: Steep orography → higher vertical velocities → enhanced relative humidity to build up condensate and thus to form MPCs.

Goals of this study

- Understand how aerosols and cloud dynamics (vertical velocity) affect droplet formation in an Alpine environment.
- Recognize under which regimes droplet formation is velocity-limited or aerosol-limited.
- Estimate the contribution of updraft velocity variance to the total variability in predicted droplet numbers.

Objectives

- This study analyzes observational data and measurements collected in February/March 2019 as part of the RACLETS field campaign in the alpine region.

- The CCN activity of the aerosol as well as their size distribution and chemical composition are discussed.

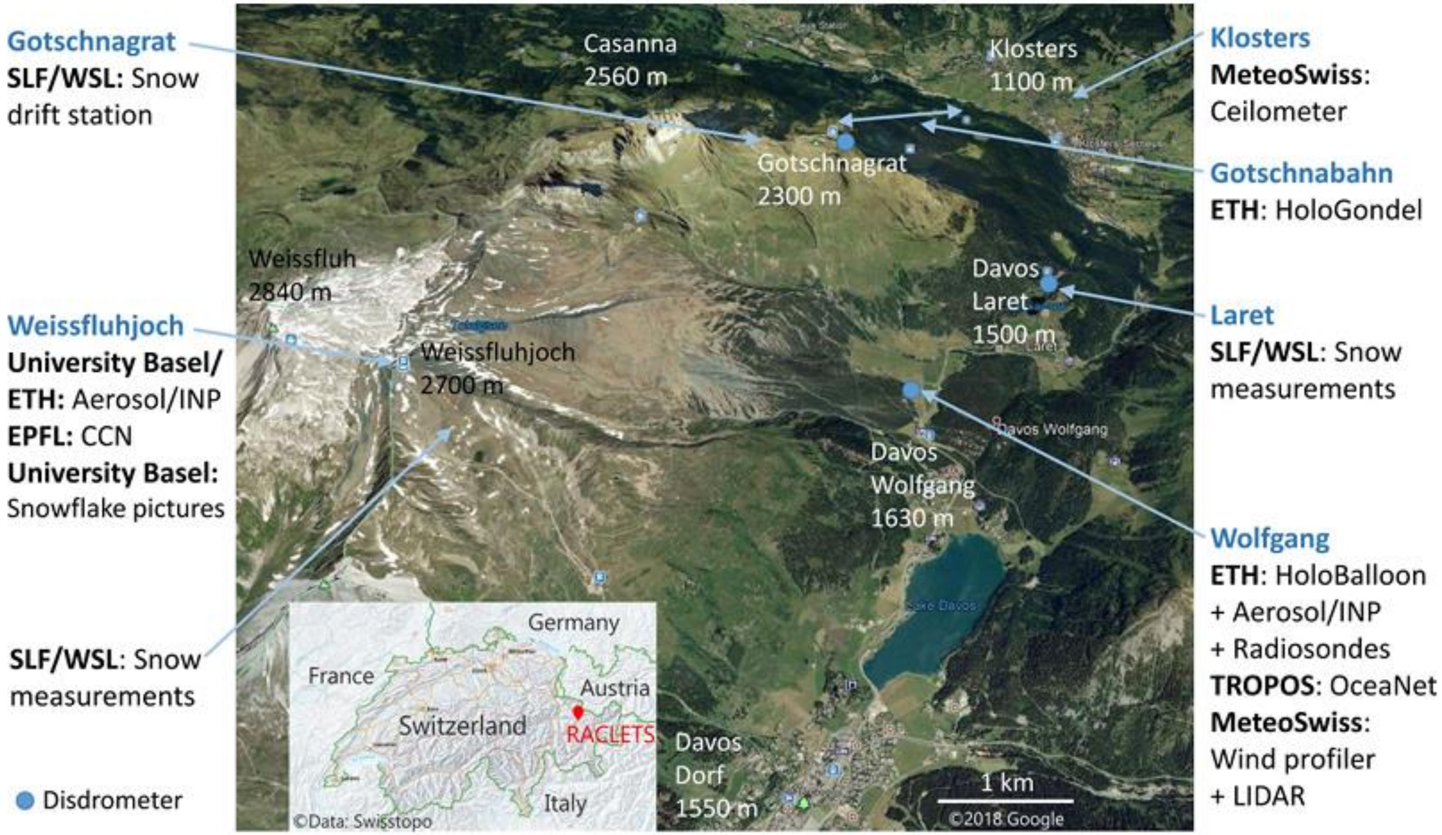
- The in-situ measurements are coupled with a state-of-the art droplet parameterization to investigate the drivers of droplet variability in the orographic mixed-phase clouds.

Field Campaign

RACLETS campaign
(**R**ole of **A**erosols and **CL**ouds **E**nhanced by **T**opography on **S**now)

Main focus of the campaign

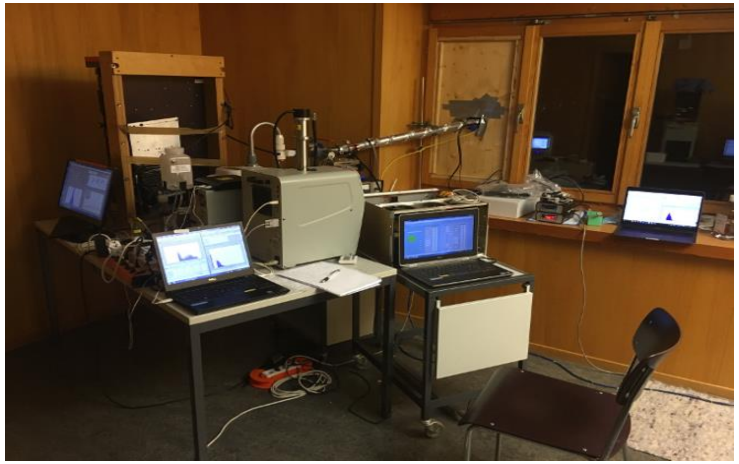
Improve the understanding of precipitation formation in clouds and snow deposition on the ground



- February-March 2019
- Davos region in Switzerland
- Includes aerosol, cloud, precipitation and snow measurements

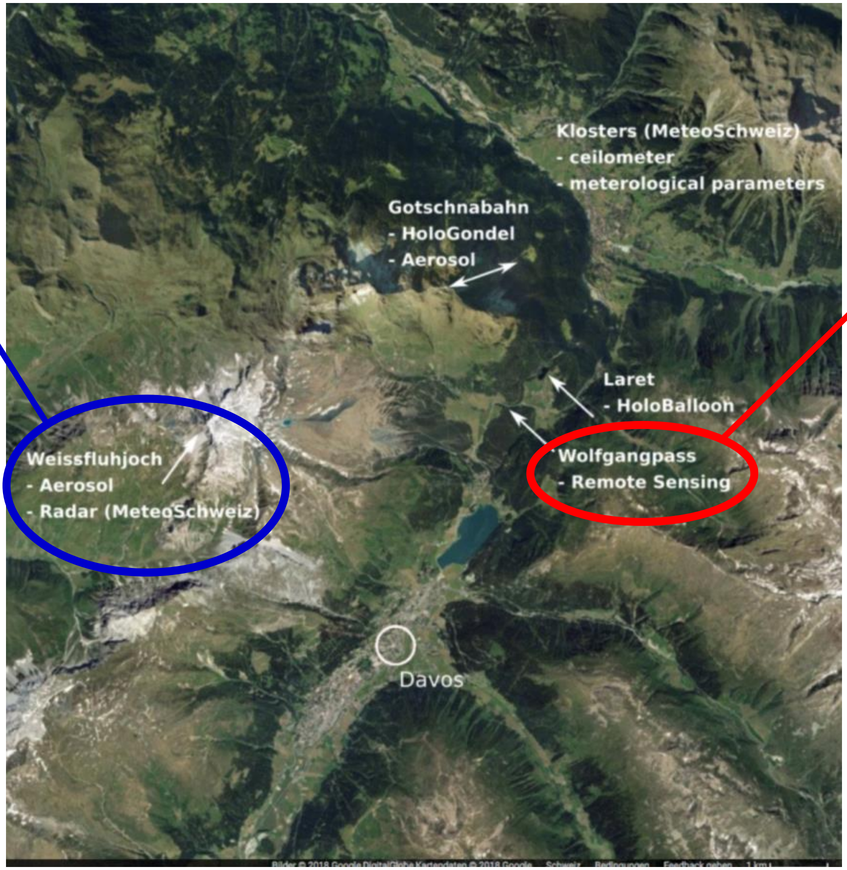
Data & Methods

Instrumentation



*The measurement site in the high-alpine research station of Weissfluhjoch (**WFJ**), 2700 m above sea level (a.s.l.)*

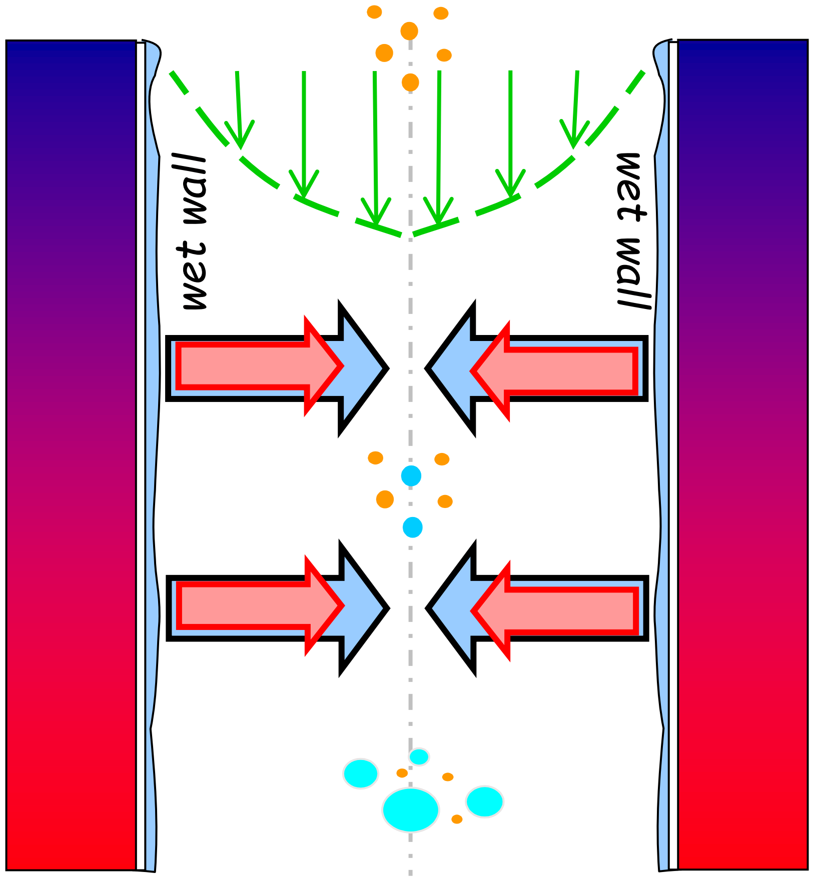
- **Measurements @WFJ :**
  - CCN measurements by a DMT CCN chamber
  - Aerosol number size distribution data by a Scanning Mobility Particle Sizer (SMPS)
  - Meteorological data available from the MeteoSwiss observation station



*The main measurement site in Davos Wolfgangpass (**WOP**), 1630 m a.s.l*

- **Measurements @WOP :**
  - SMPS aerosol number size distribution data
  - Wind measurements by a mobile wind profiler of MeteoSwiss

CCN Measurements & Sampling Strategy



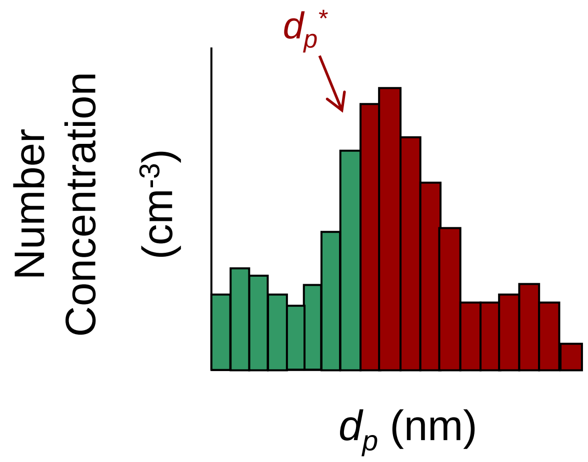
- Metal cylinder with wetted walls
- Streamwise Temperature Gradient
- Water diffuses faster than heat
- Supersaturation, S, generated at the centerline = f(Flowrate, Pressure and Temperature Gradient)
- Particles that activate to form droplets are counted as CCN and sized by an optical particle counter
- Products: CCN concentrations at six S between 0.1 to 0.8 %
- Cycle considers 10 minutes at each supersaturation → CCN spectrum every hour
- When switching S, instrument transients affect measurements, so they are "filtered" out

Roberts and Nenes (2005); Moore and Nenes (2010)

Inferring particle hygroscopicity parameter (κ)

1. Measure CCN concentration, [CCN], at a given *SS%*, this can be done in either constant flow or scanning flow instrument modes

2. Find where backwards integrated size distribution = [CCN] to obtain the critical diameter, d_p^*



3. Use κ-Köhler theory to calculate κ:

$$\kappa \approx \frac{4A^3}{27 d_p^3 S^{*2}}$$

$$A = 4M_w \sigma_w / RT\rho_w$$

κ ~ 1 for seasalt, ~ 0.6 for $(NH_4)_2SO_4$, ~ 0.1-0.2 for BB
a "proxy" for chemical composition

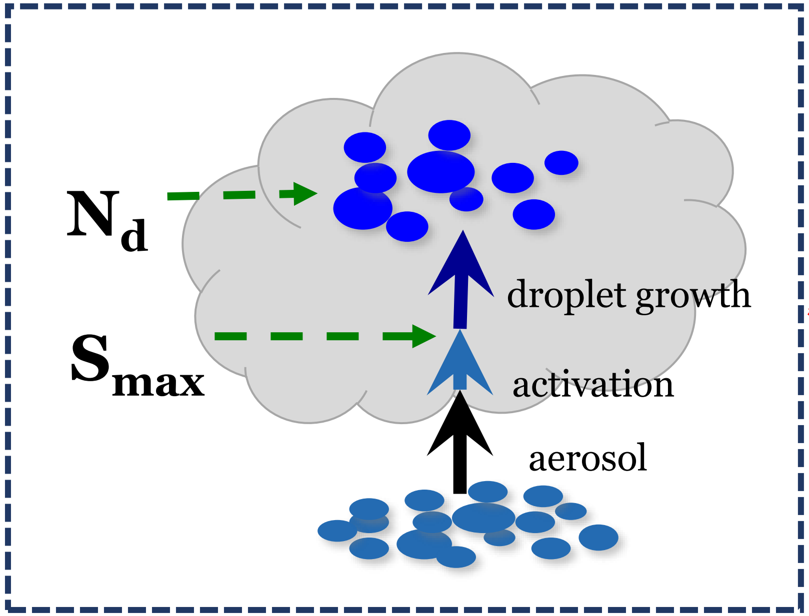
Petters and Kreidenweis (2007); Moore et al. (2012); Bougiatioti et al. (2016)

Droplet Activation Parameterization

Observed aerosol size distribution

Observed aerosol composition

Observed cloud updraft velocity



Maximum in-cloud Supersaturation (Smax)

Predicted Cloud Droplet Number (Nd)

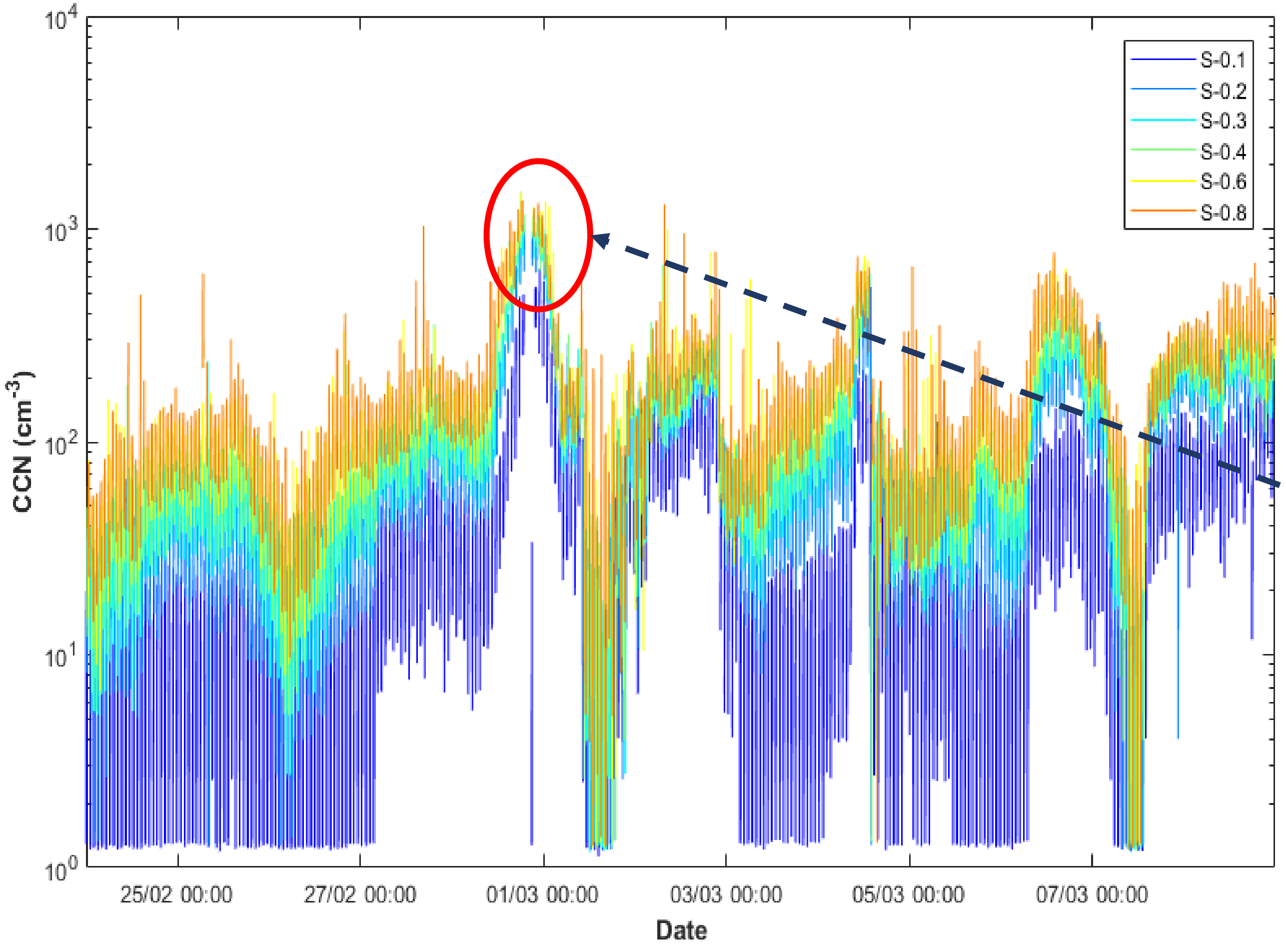
Nenes and Seinfeld (2003); Fountoukis and Nenes (2005); Morales and Nenes (2014)

Results & Discussion

Measured CCN concentrations at the mountain site WFJ

Period of Interest: 24.02.2019 – 08.03.2019

- CCN concentrations ↑ as supersaturation (S) ↑ (as expected)
- Relatively low CCN concentrations even at the highest S → representative of a remote continental measurement site

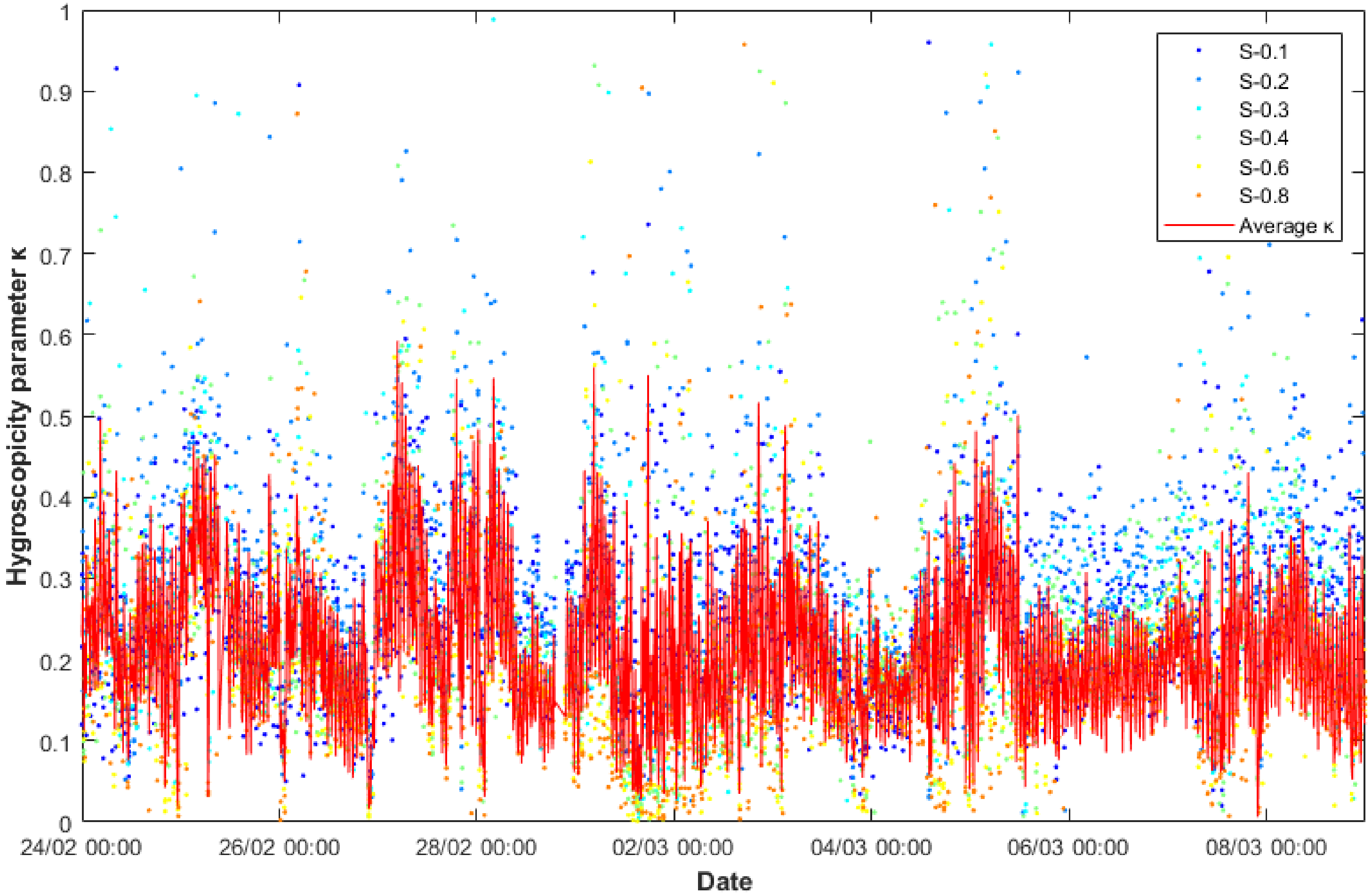


Sudden and short-lived fluctuations in the CCN concentrations could be related to meteorological transport processes (e.g. large-scale synoptic flow, vertical transportation)

CCN number concentrations measured at 6 different supersaturations (0.1-0.8%)

CCN-derived κ-parameter at WFJ

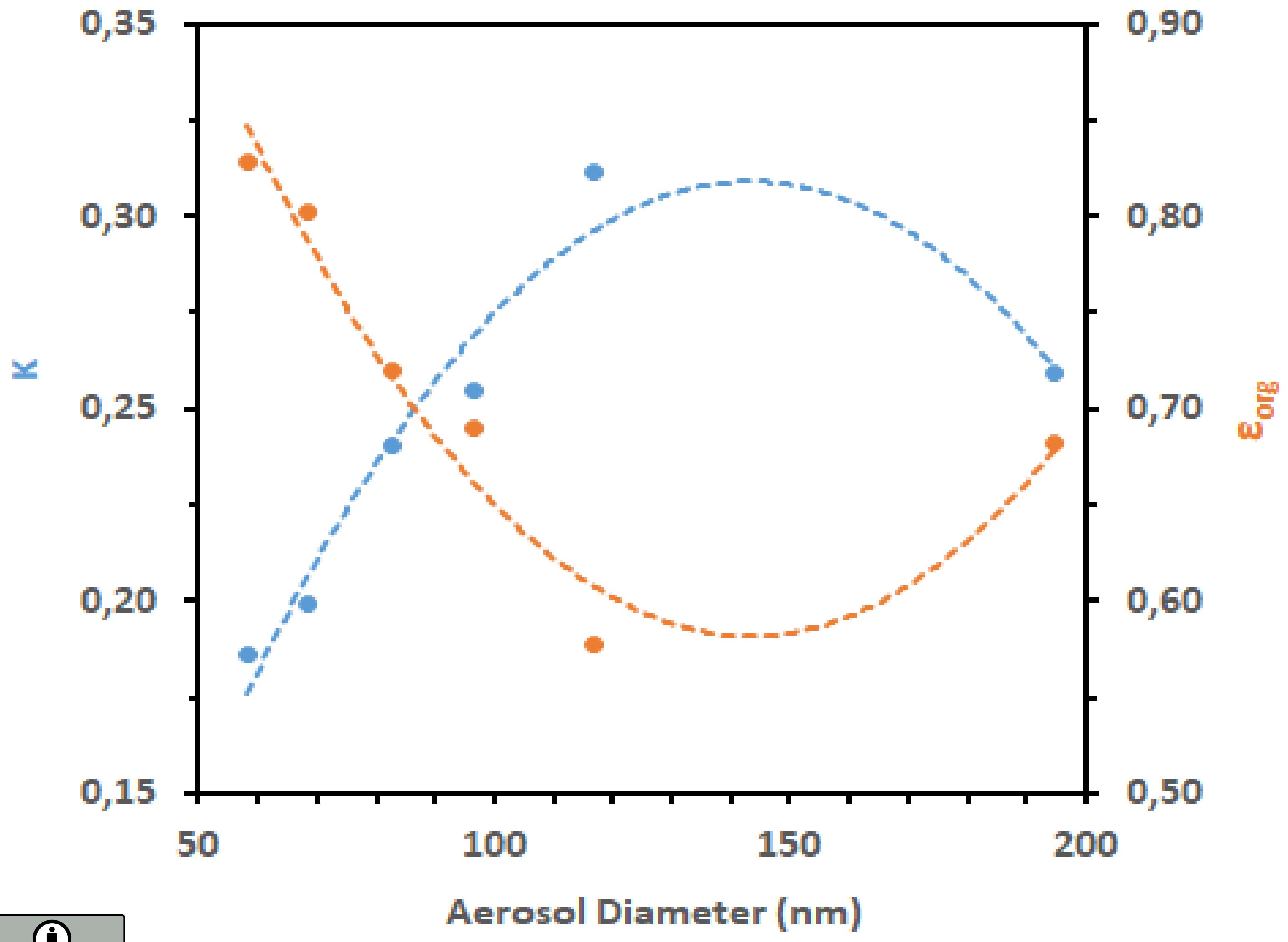
Hygroscopicity parameter κ wraps all the chemical complexity of particles → it reflects particles composition



60% drop in κ-parameter as the particles get smaller (i.e., with higher supersaturation) → indication of enrichment by organics

Supersaturation (%)	κmean ± std
0.1	0.26 ± 0.10
0.2	0.31 ± 0.13
0.3	0.25 ± 0.13
0.4	0.24 ± 0.13
0.6	0.20 ± 0.12
0.8	0.19 ± 0.11

Size-resolved κ-parameter



ε_{org} (organic mass fraction) assuming a mixture of an organic and inorganic component with characteristic κ_{org} ∽0.1 and κ_{inorg} ∽0.6

- Aged particles (>100 nm) are more hygroscopic than the smaller ones
- Sub-100 nm particles are enriched in organic material – BB influence?
- κ ∽ 0.2 - 0.3, typical of continental aerosol

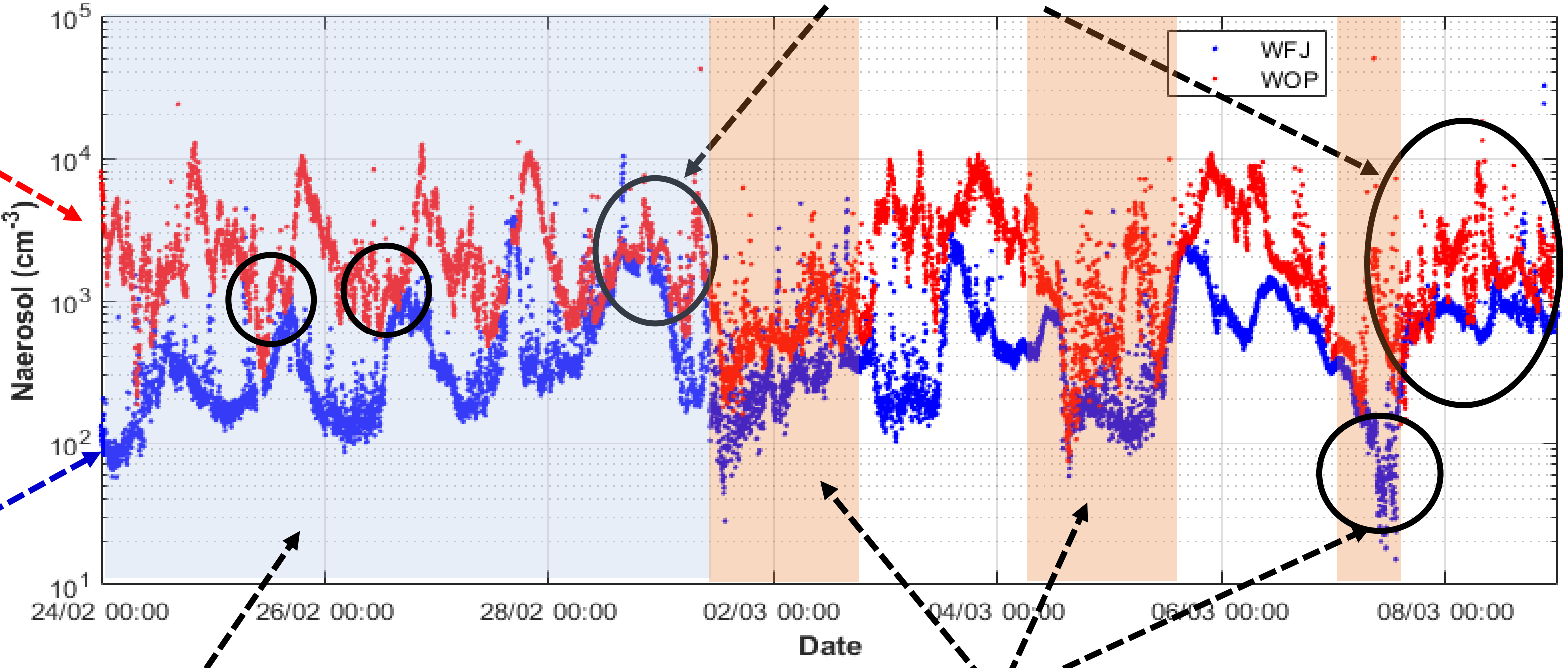
Timeseries of total aerosol number (SMPS)

Question: *What are the potential differences in aerosol variations between valley measurements at WOP and measurements taken at high-altitude stations like WFJ?*

WOP timeseries -> increased aerosol concentrations compared to WFJ
- Local aerosol sources (biomass burning?)

WFJ timeseries -> Continental background site because of its altitude and location
- Long-range transport

Similar aerosol concentrations observed at both sites → same air masses?

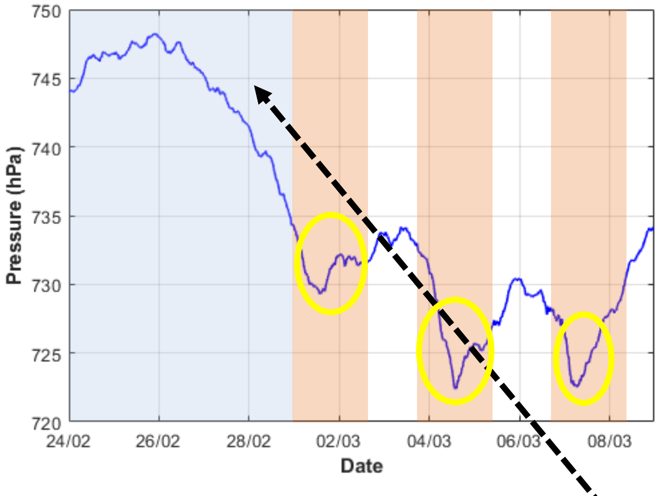


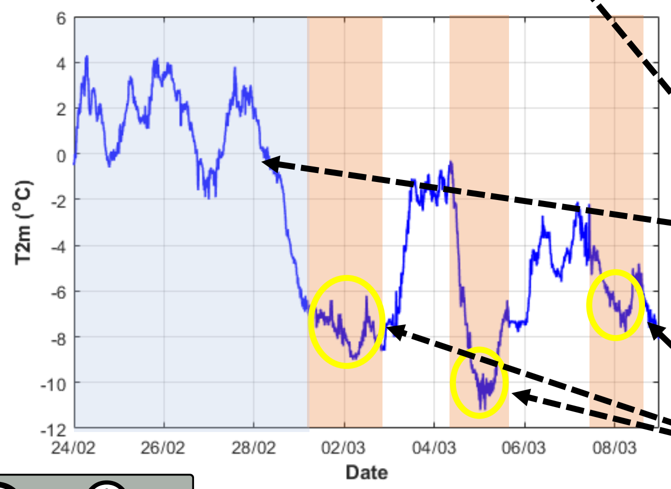
Diurnal cycles between both sites anticorrelate. Mountain BL dynamics? Midday: when two sites meet - experience same air masses?

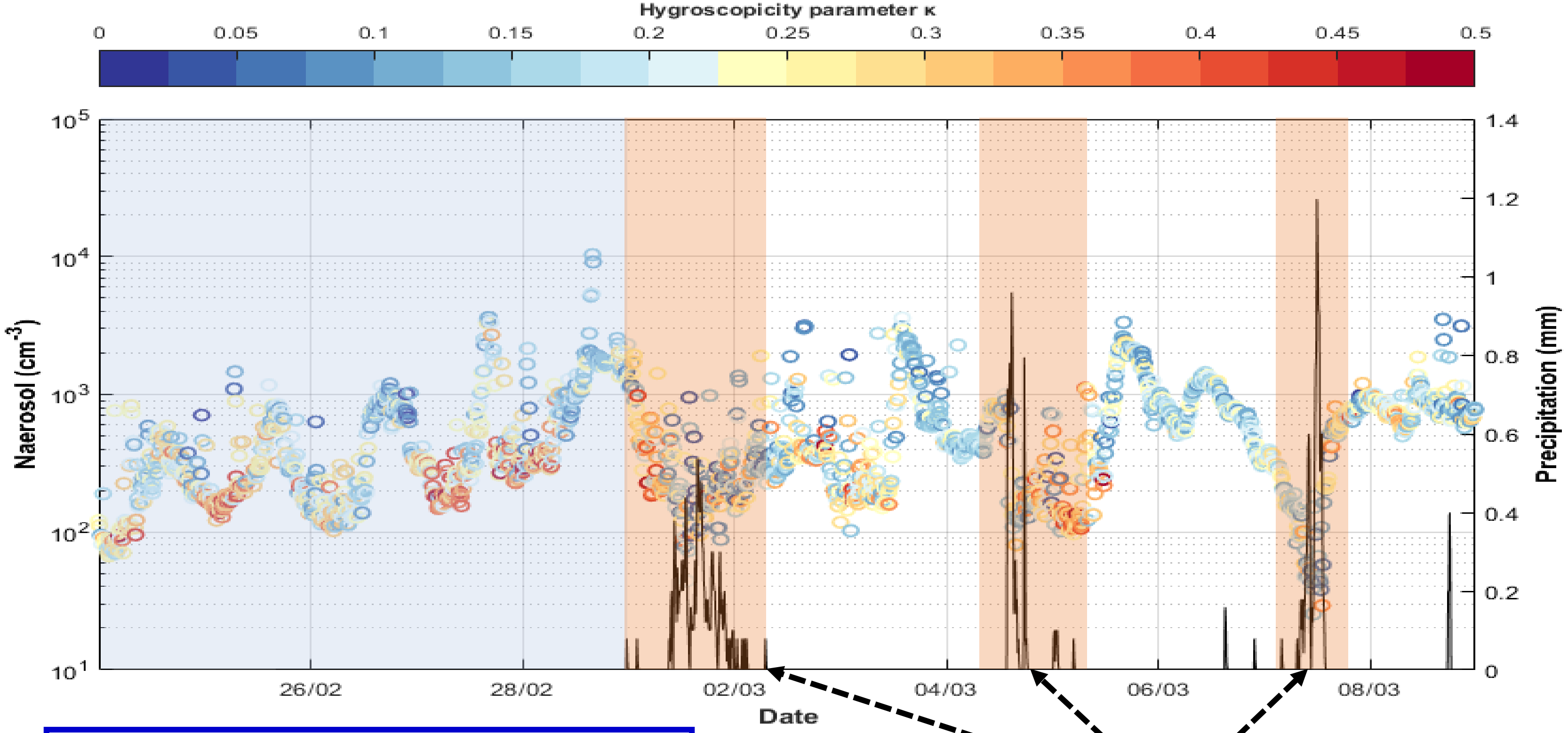
Snow? Both sites experience the same (low) aerosol levels.

Timeseries of total aerosol number (SMPS)

Meteorological Data by MeteoSwiss weather station at WFJ







High pressure and temperature observed at station level → fair weather until 28.02

Sharp pressure and temperature decrease → cold front

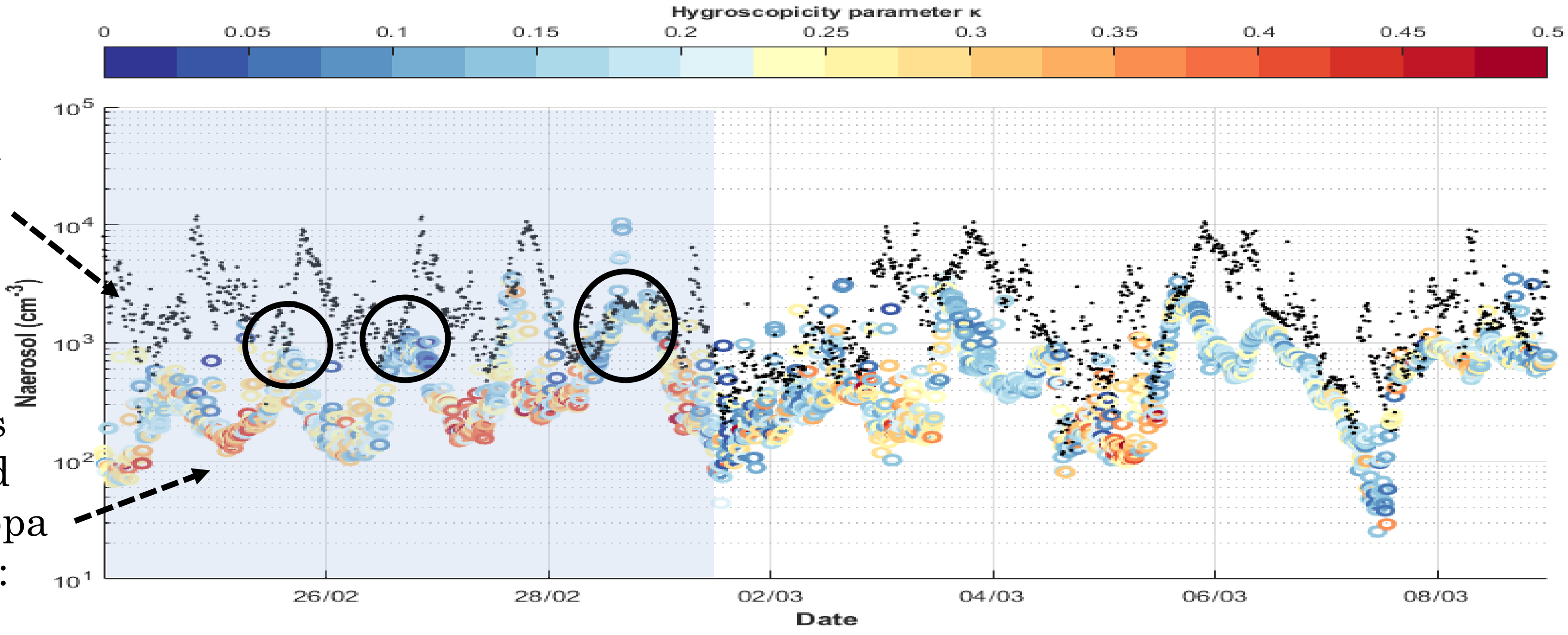
Aerosol concentration drop coincides with increased measured precipitation at WFJ → wet deposition of more hygroscopic (more aged) aerosols

Timeseries of total aerosol number (SMPS)

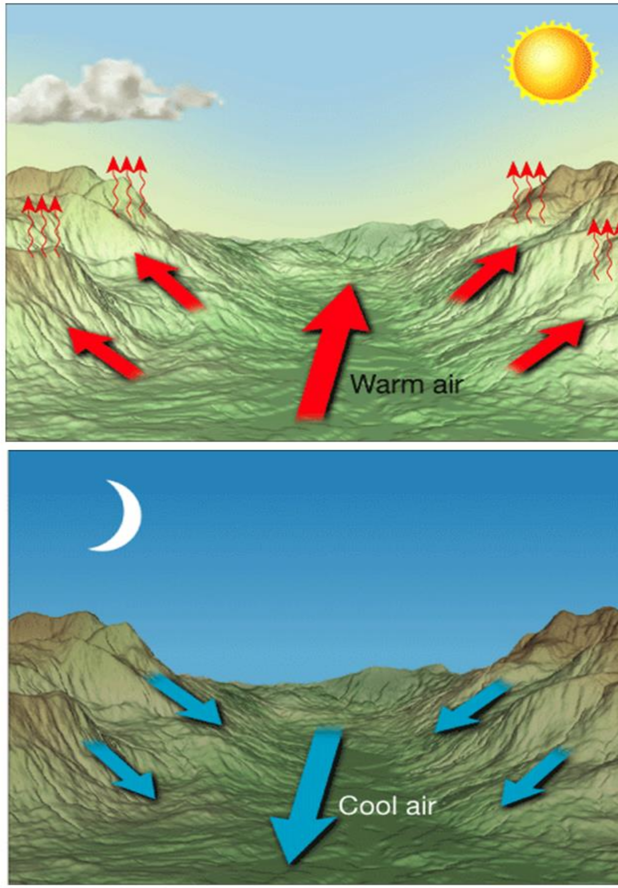
Question: *Can boundary layer dynamics explain the diurnal cycles seen during the first half of the period of interest?*

Black dots: **WOP**

Circles colored by kappa values: **WFJ**

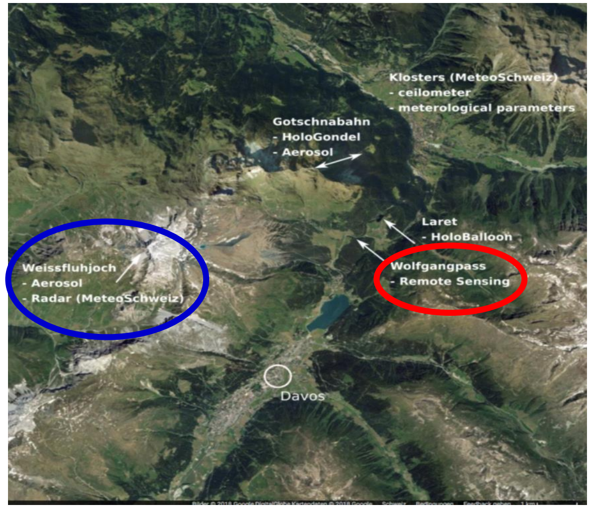


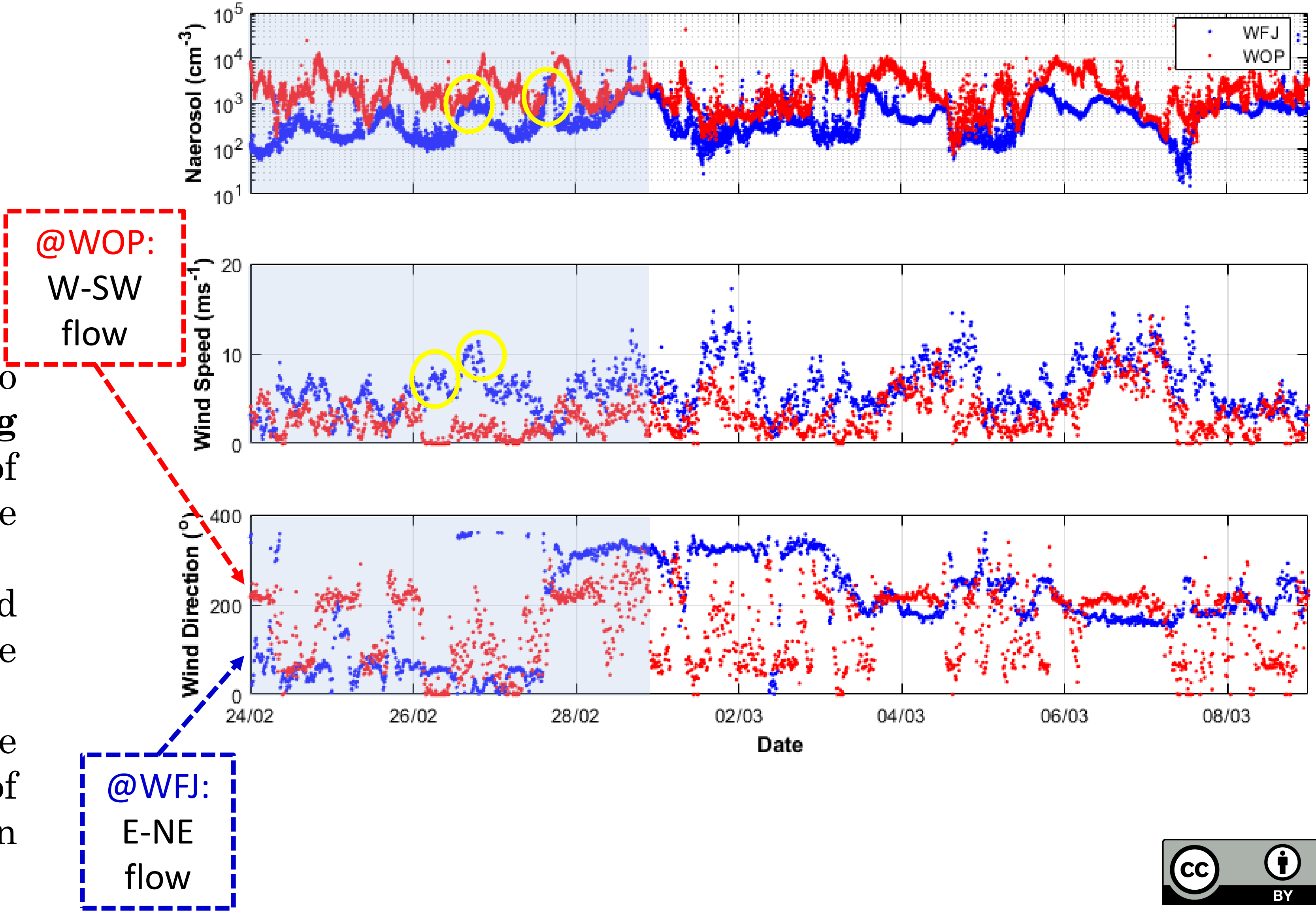
- **Daytime**: upslope flow due to **thermal convection** → air in the boundary layer of WOP rises up the slope increasing the concentrations of less hygroscopic (less aged) aerosols observed during afternoon at WFJ (black circles)
- **Evening**: the situation reverses, concentration max @WOP and WFJ influenced by FT air (lower concentrations of more hygroscopic aerosols)



The up- and downslope flows produced by inclined cold or warm boundary layers that form above the slopes.

Timeseries of total aerosol number (SMPS)





- Upslope flow due to **mechanically forced lifting** caused by the deflection of strong winds (~10 m/s) by the mountain slope?
- The wind direction measured @WFJ coincides with the relative location of WOP site.
- The steep orography over the Alps might transform part of this strong horizontal motion into vertical motion.

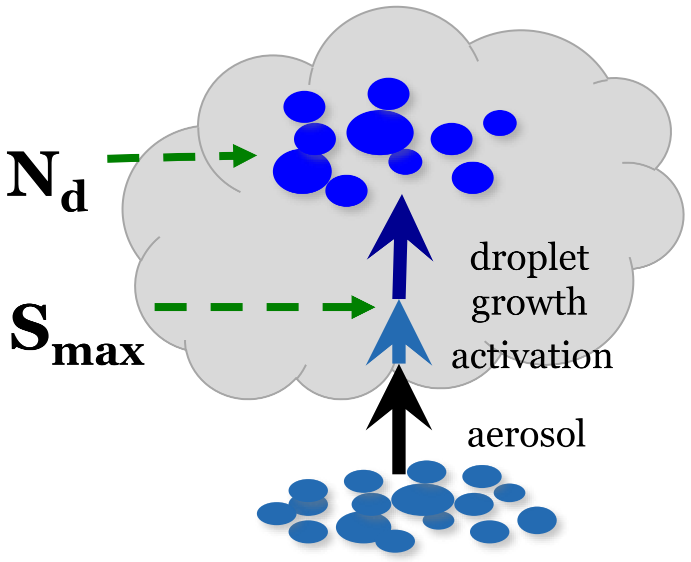
From Aerosol to Droplets

- **We use** Morales and Nenes (2014) droplet formation parameterization, with sensitivities calculated from numerical adjoints, etc. to determine s_{max}, and cloud susceptibility to aerosol and vertical velocity.

INPUT: P,T, vertical winds (σ_w), aerosol size distribution+κ

OUTPUT: N_d ("potential"), S_{max}, $\partial N_d/\partial N_a$, $\partial N_d/\partial \sigma_w$, $\partial N_d/\partial \kappa$

- **Droplet numbers and sensitivities shown are the PDF-averaged value (**integrated over the positive part of the vertical velocity spectrum**).**
- **We don't know σw, so we do a sensitivity calculation for σw=0.1-0.6 m s^{-1}**
- In-cloud supersaturation for most of the simulations is around 0.1-0.3% → **κ=0.25** to run the droplet parameterization.
- Same value of κ used for WOP



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0.8	0.19 ± 0.11

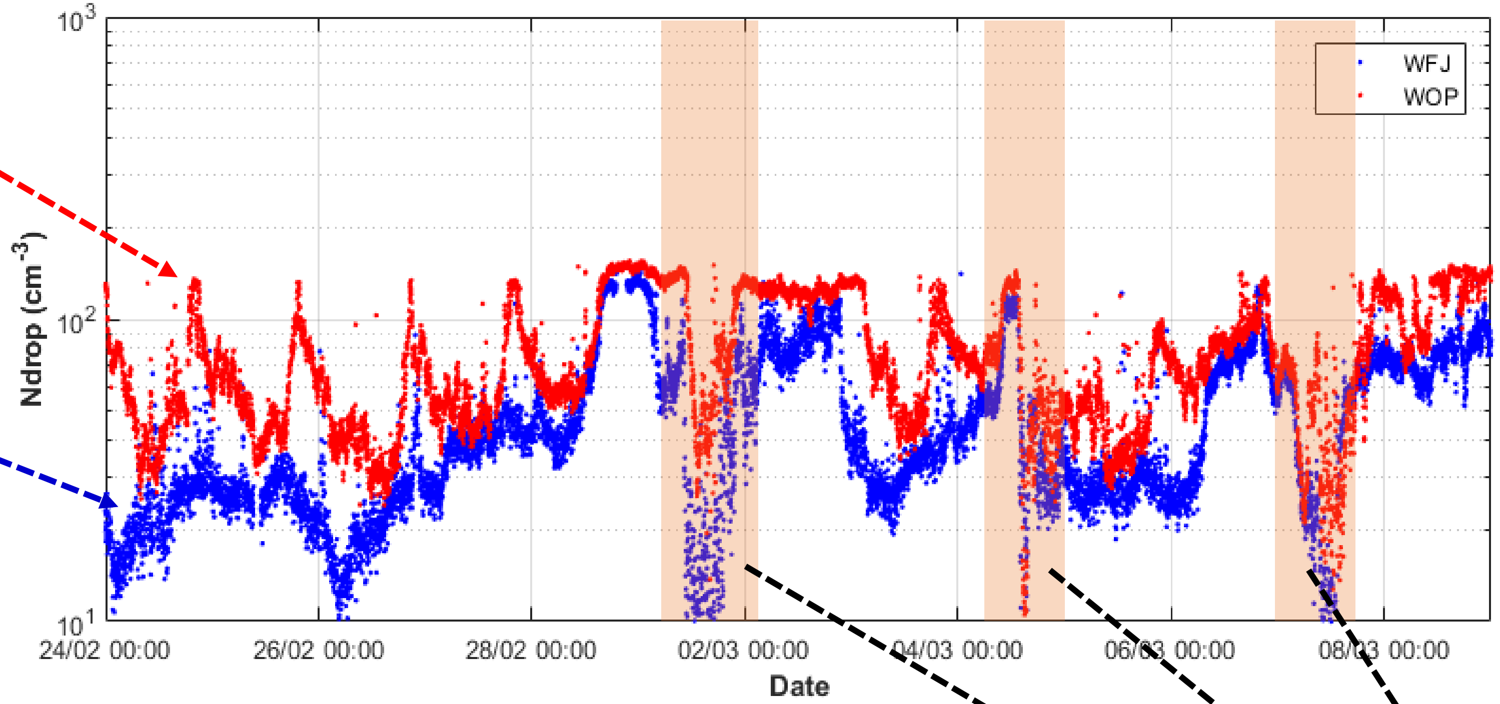
Potential droplet timeseries (WFJ,WOP) (σ_w=0.1ms^{-1})

Cloud droplet concentrations in WOP are ∽ 10 x more than in WFJ

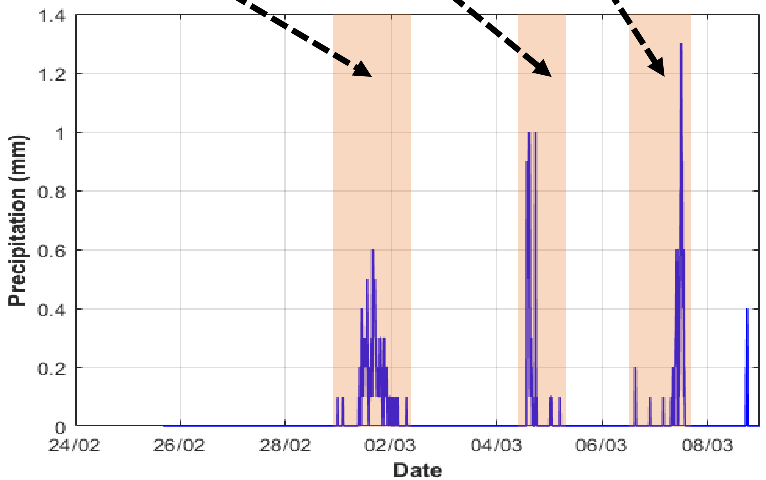
Pronounced diurnal cycle in WOP, no cycle in WFJ (contrasts SMPS data)

Why?

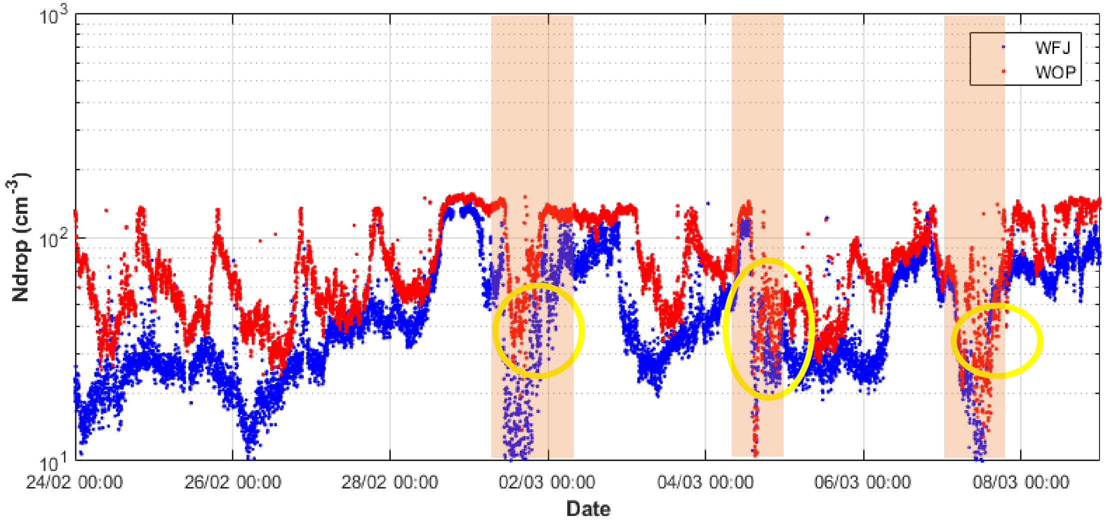
- Aerosol particles brought up from below may be enriched in particles too small to activate into droplets
- Accumulation mode aerosols that activate may be more regional (aged)



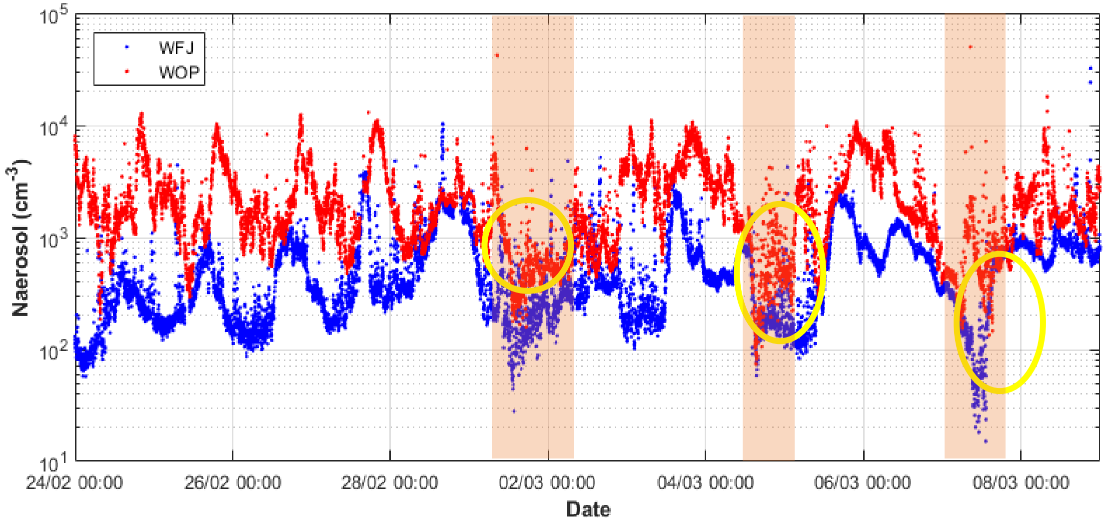
Precipitation is again the “culprit” behind the low cloud droplet concentrations observed at WFJ where Ndrop → 0

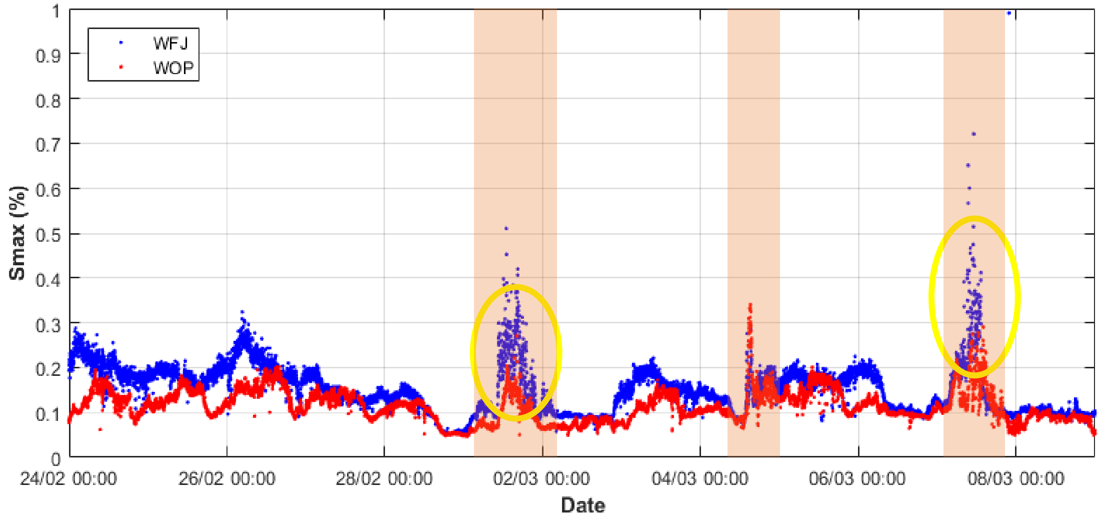


Potential droplet timeseries (WFJ,WOP) (σ_w=0.1ms^{-1})

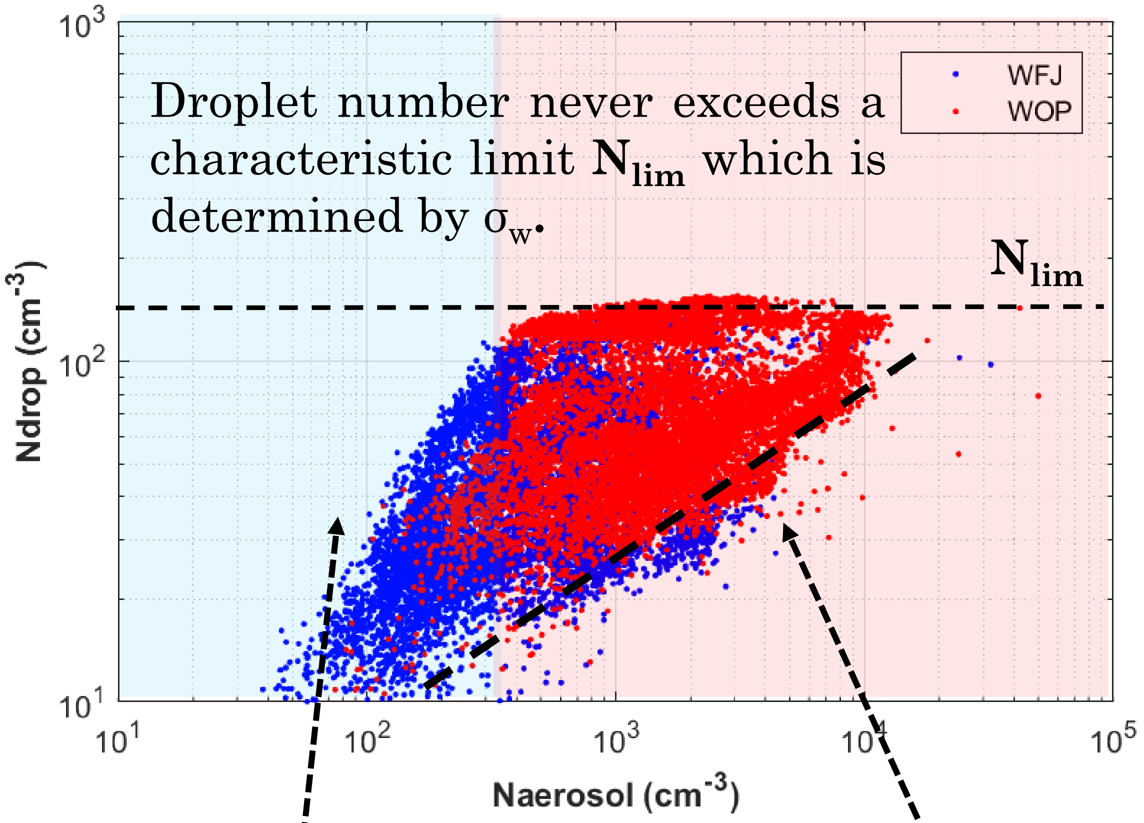


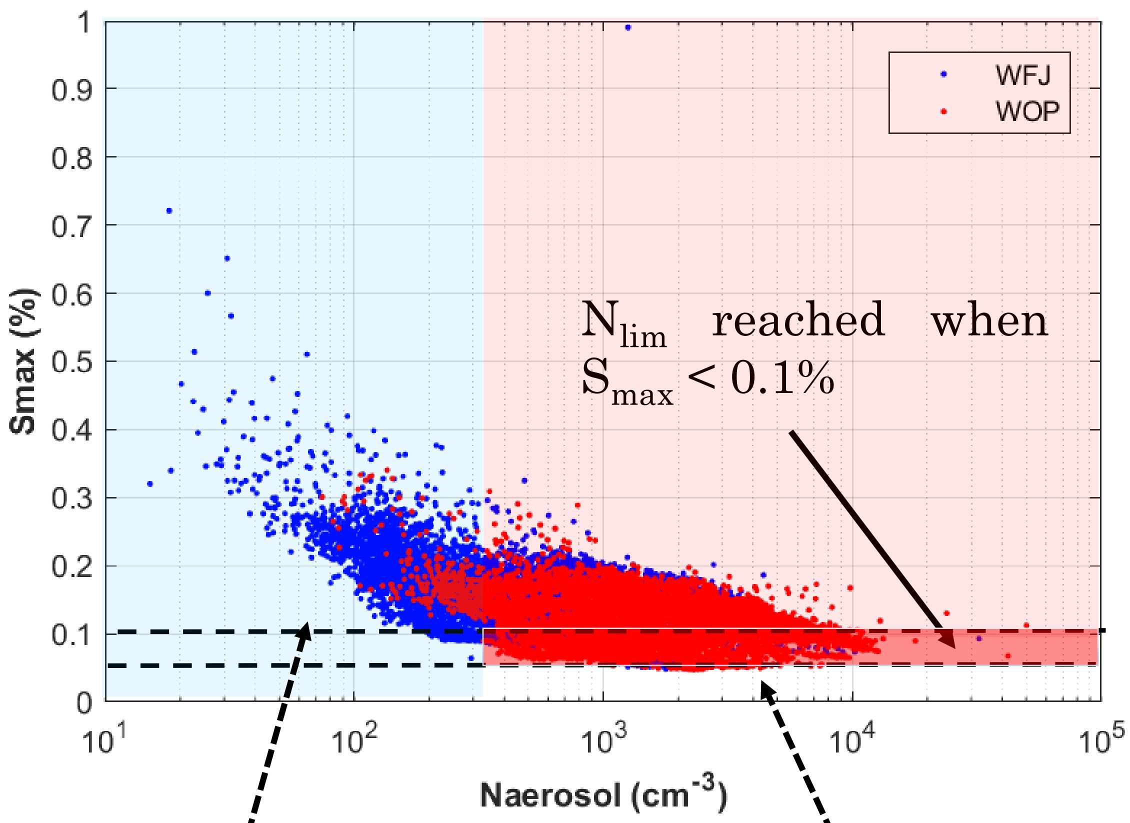
- Significant **drop in N_d** on **01.03**, **05.03** and **07.03** (yellow circles) coincides with **high S_{max}** → **few CCN** (~10 cm^{-3}) up to 0.4-0.5% supersaturation.
- 01.03, 05.03 events, $N_{aerosol}$ "high" (100-300 cm^{-3}) at both WOP/WFJ → small particles that activate above 0.3-0.5%.
- This is not seen in 07.03 for WFJ





Summary of droplet response to changes in total aerosol concentration (σ_w=0.1ms^{-1})





N_{drop} never "hits" N_{lim} for $N_{aerosol}$ < 300cm^{-3}, we are always in the "**aerosol limited**" regime.

When water-vapor competition effects are strongest, N_{drop} is **velocity-limited**.

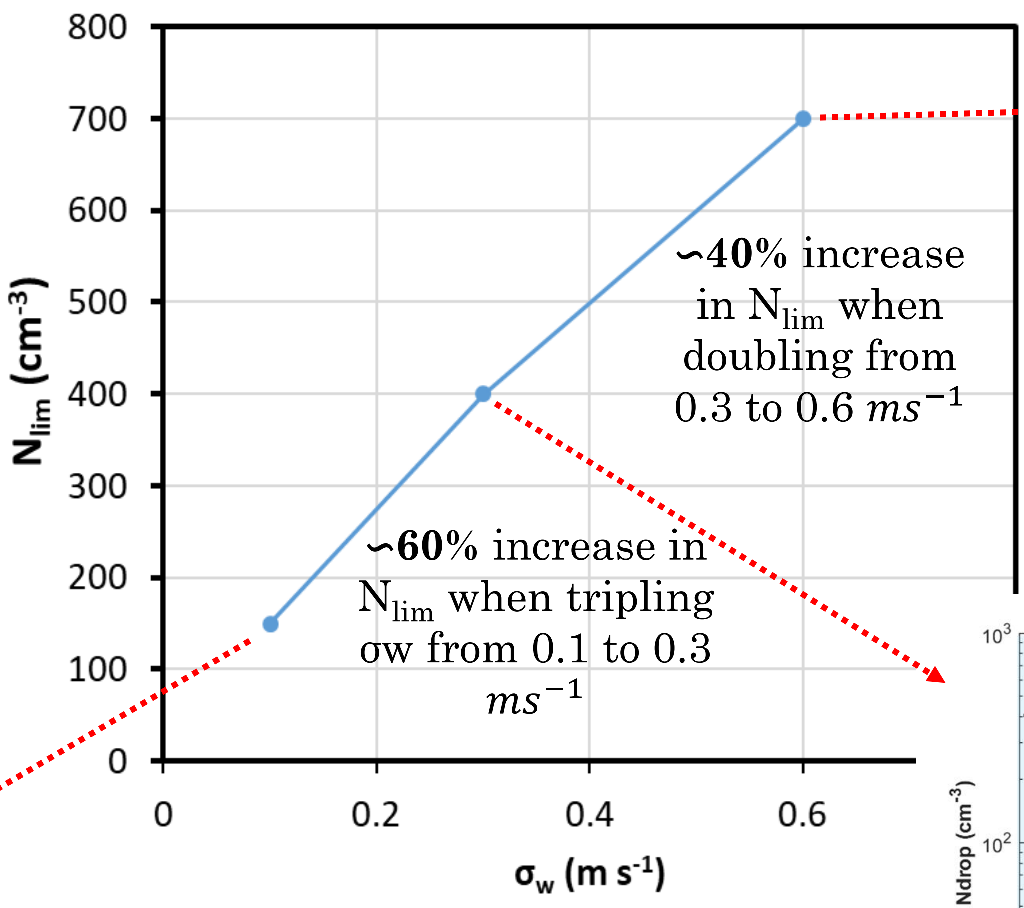
Within the aerosol-limited regime Smax values are large enough to activate almost all of the particles except for the very small ones.

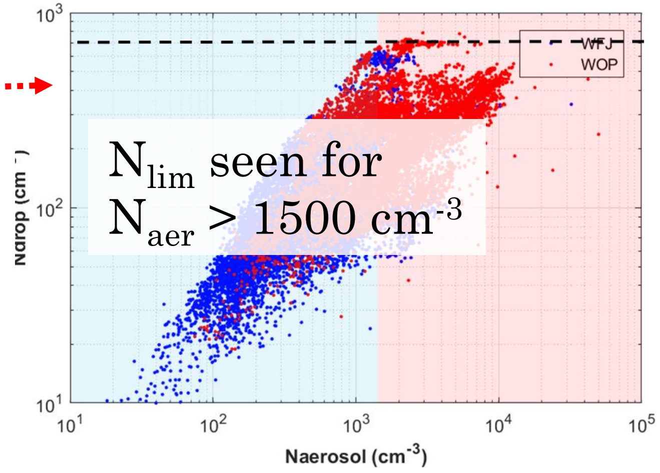
Within the velocity-limited regime the Smax values are very low → only a few particles are able to activate to cloud droplets.

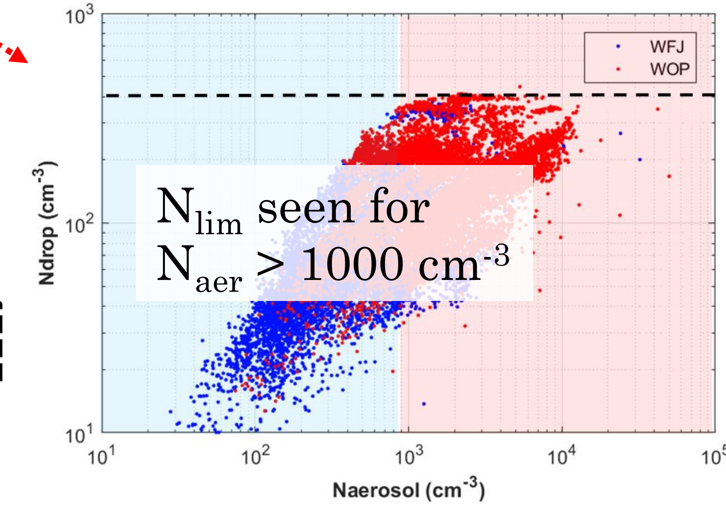
N_{lim} is a reflection of the dynamics

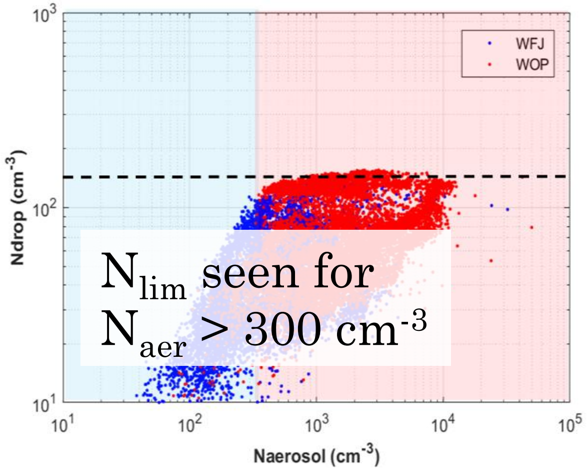
Question: *What is the impact of increased updraft velocity on the limiting droplet number N_{lim}?*

- When boundary layer turbulence is low (σ_w<0.1 ms^{-1}) → aerosol variability does not result in a significant change in N_{lim}.
- In a more convective boundary layer (σ_w≥0.3 ms^{-1}), when aerosol levels increases → the impact on N_{lim} is more profound.









As $\sigma_w \uparrow N_{lim} \uparrow$ since Supersaturation ↑

Conclusions

Some take-home messages

- CCN-derived κ ∽ 0.2 - 0.3 → typical of **continental aerosol**
- Accumulation mode particles (~100nm diameter) are **more hygroscopic** than the smaller ones (~50nm diameter), likely from an enrichment in organic material.

- **Droplet formation for σ_w =0.1 ms^{-1}**: always aerosol limited if N_{aer} < 300 cm^{-3} and velocity-limited when S_{max} drops below 0.1%. Droplet number never exceeds the limit N_{lim} ~ 150 cm^{-3}.
- **At σ_w =0.3, 0.6 ms^{-1}**, same behavior is seen, but the aerosol limited regime is extended to N_{aer} < 1000, 1500 cm^{-3} respectively.
- N_{lim} responds **proportionally** to changes in σ_w.
- When in the aerosol-limited regime, droplet number formation is driven by aerosol variability.
- When velocity-limited, droplet number formation is driven by σ_w variability.

Lausanne

Thank you for your interest!

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