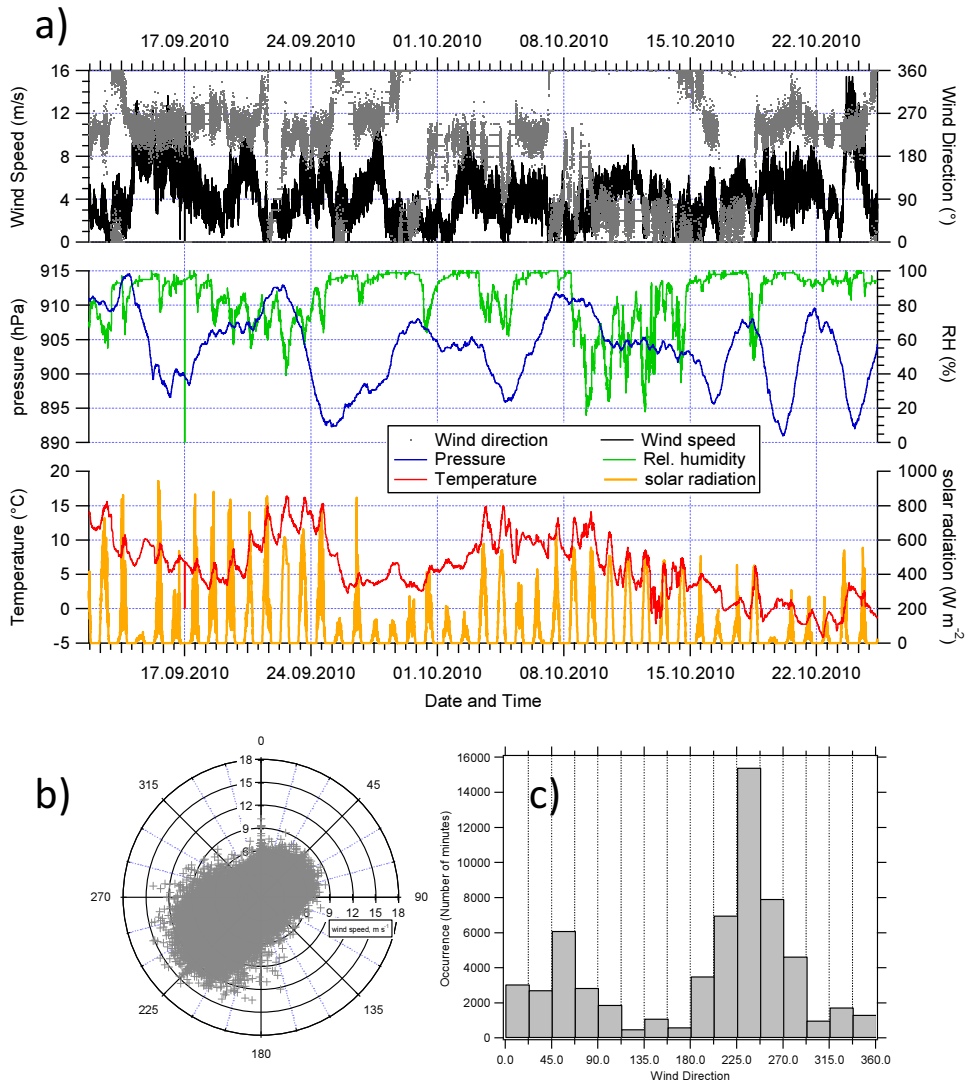


Table S1: Relative ionization efficiencies (RIE) used in this work. RIE_{NH_4} and RIE_{SO_4} were inferred from calibrations during this field study. RIE_{NO_3} , RIE_{org} , and RIE_{Cl} are default values from published laboratory measurements or field intercomparisons (Alfarra et al., 2004; Takegawa et al., 2005; Canagaratna et al., 2007).

Instrument	RIE_{NO_3}	RIE_{NH_4}	RIE_{SO_4}	RIE_{Org}	RIE_{Cl}
C-ToF-AMS	1.1	4.11	0.70	1.4	1.3
HR-ToF-AMS	1.1	3.74	1.49	1.4	1.3



Figure S1: Photographs of the summit site. Left: Measurement building and tower; middle: instruments on the 20 m tower; right: CVI and interstitial inlet at ca. 15 m altitude, facing south-west (215°).



5 **Figure S2. Overview of the meteorological parameters measured by the Vantage Pro weather station on the tower at the summit site. a) time series of relevant parameters; b) polar graph with wind speeds and wind directions; c) histogram of wind directions.**

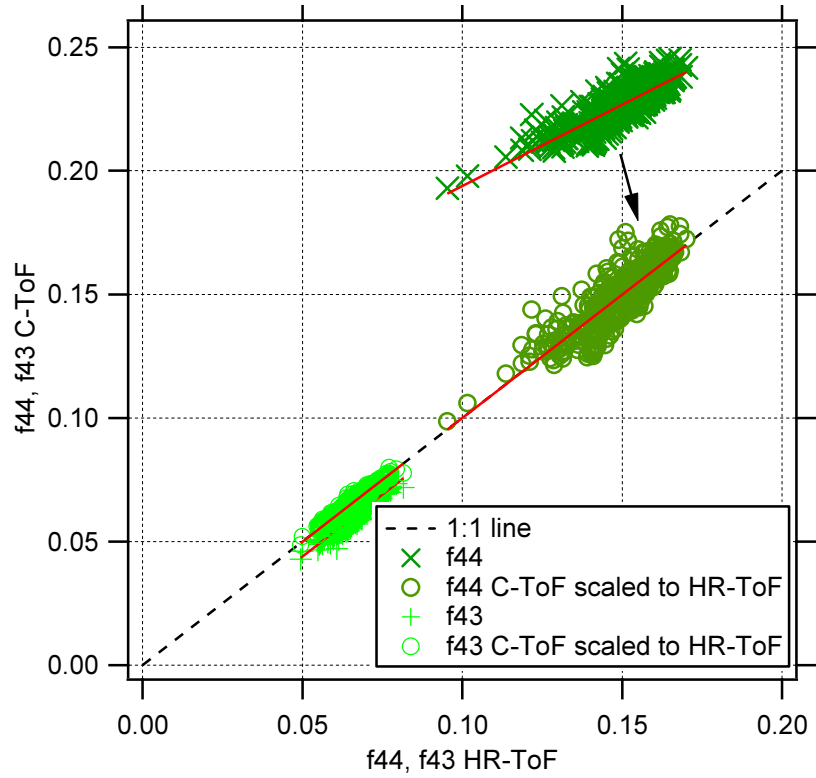


Figure S3. Scaling of f_{44} (dark green) from C-ToF-AMS to HR-ToF-AMS. For f_{43} (light green), the scaling was only marginal. This scaling is based on the cloud-free intercomparison period from September 19 to 25. The red lines are linear fits to the data.

5

Original values:

$$f_{44,C-ToF} \text{ vs } f_{44,HR-ToF}: \text{ slope} = 0.660, \text{ offset} = 0.128, r^2 = 0.75$$

$$f_{43,C-ToF} \text{ vs } f_{43,HR-ToF}: \text{ slope} = 1.003, \text{ offset} = -0.006, r^2 = 0.91.$$

10 Scaling equations:

$$f_{44,C-ToF,corr} = (f_{44,C-ToF,measured} - 0.128) \times 1.514 \quad (S1)$$

$$f_{43,C-ToF,corr} = (f_{43,C-ToF,measured} + 0.006) \times 0.997 \quad (S2)$$

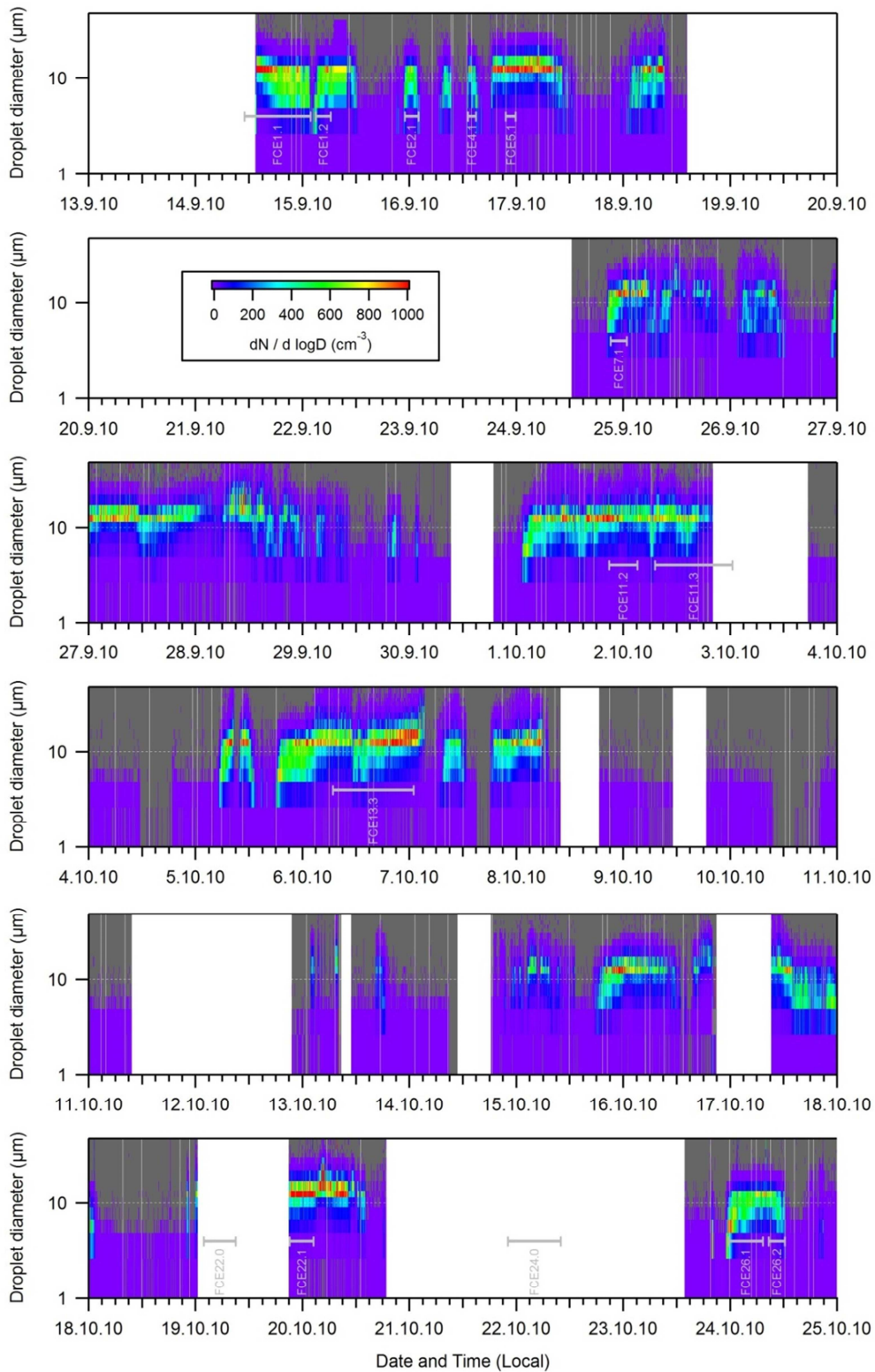


Figure S4: Time series of cloud droplet size distributions measured during HCCT-2010. The full cloud events as defined in Tilgner et al. (2014) are indicated. During FCE22.0 and FCE24.0 the FSSP was not operated due to icing at the instrument tip.

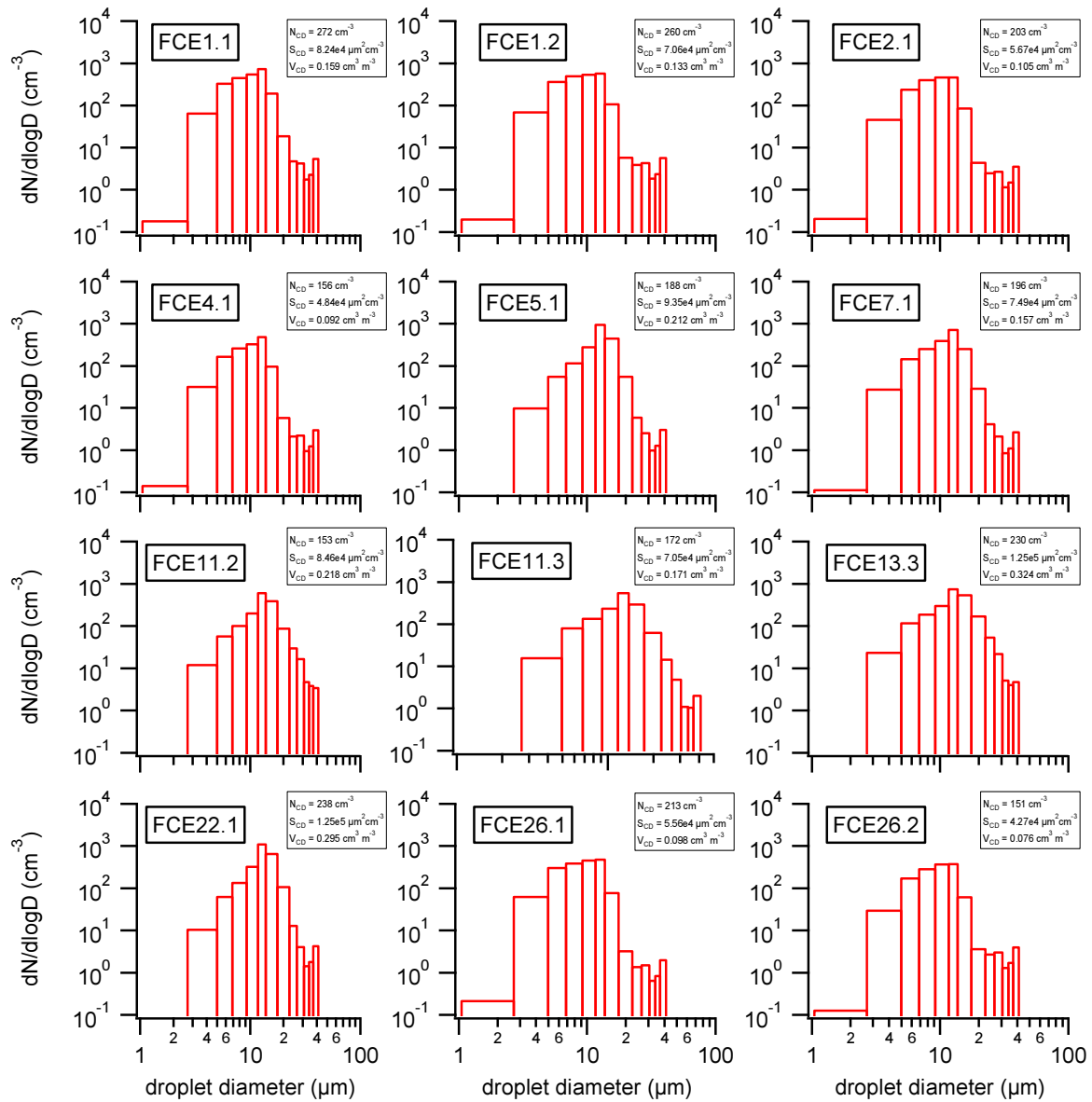


Figure S5: Number size distribution of cloud droplets measured by the FSSP-100 for all full cloud events (FCE) as given in Table 1 of the main text. The averaging times correspond to those given in Table 1, except for FCE1.1, where the first 2 hours and 23 minutes are missing, and 11.3, where the last 4h 21m are missing. During FCE22.0 and FCE24.0 the FSSP was not operated due to icing at the instrument tip. Averaged parameters are given in the legend (N_{CD} : Cloud droplet number concentration, S_{CD} : Cloud droplet surface concentration, V_{CD} : Cloud droplet volume concentration).

5

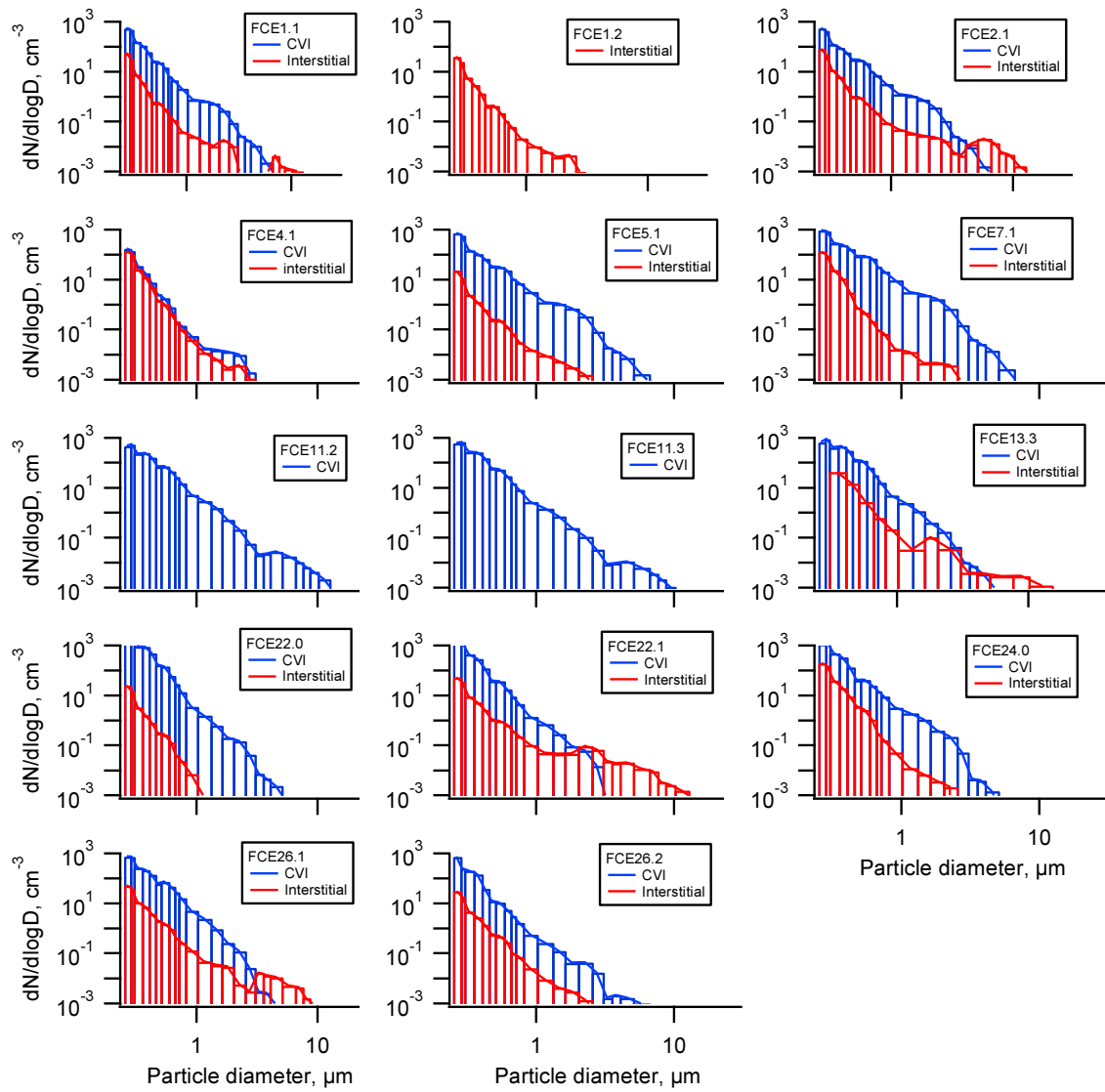


Figure S6: Size distributions for all full cloud events (FCE) measured with the optical particle counters (model Grimm 1.109; only the interstitial data during FCE 13.3 were measured by a Grimm 1.108). Missing data (FCE1.2, FCE11.2 and 11.2) are due to instrumental failures. CVI data were corrected for sampling efficiency and enrichment.

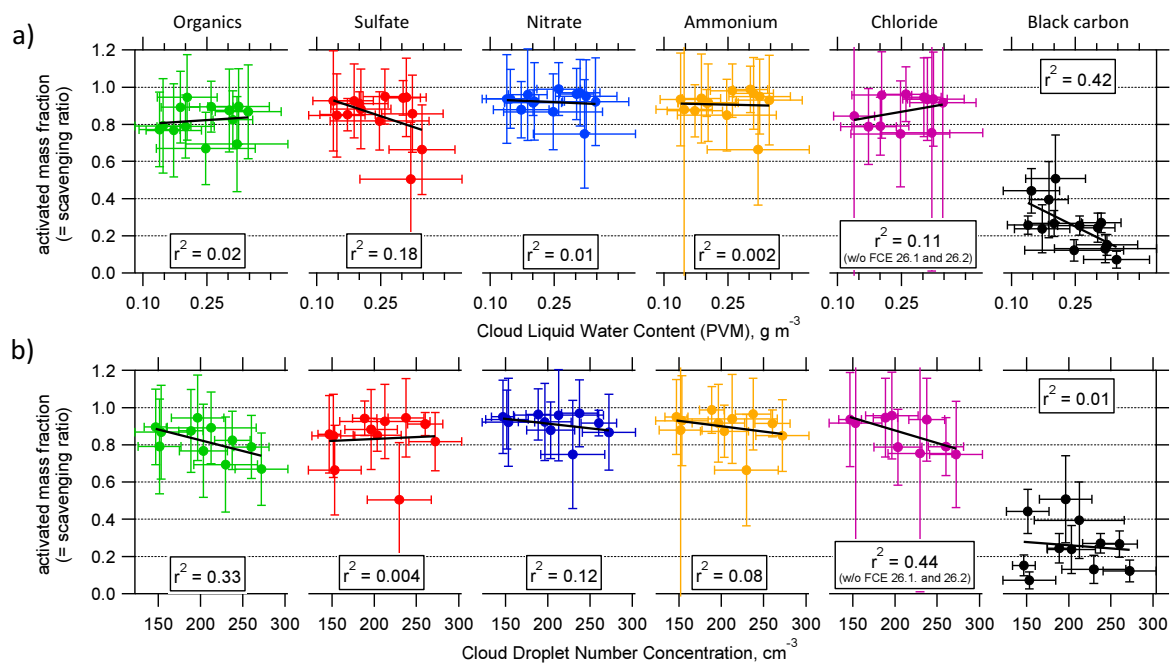


Figure S7: Scavenging efficiency as a function of liquid water content (upper panel) and of cloud droplet number concentration (lower panel). Linear regression lines were fitted to the data and correlation coefficients are given. For FCE 22.0 and 24.0 no FSSP data are available. There is almost no correlation between the scavenging efficiency and LWC. Chloride shows a positive linear correlation with a correlation coefficient (r^2) of 0.11, whereas sulfate and black carbon show negative correlations with $r^2 = 0.18$ and 0.42 , respectively. It has to be noted the FCE26.1 and FCE26.2 were not used for chloride because the mass concentrations were below the detection limit. Especially the negative correlation of SE_{sulfate} with LWC is in contradiction to observations by Kasper-Giebl et al. (2000) who found a constant increase of SE_{sulfate} with LWC. Also the SE values show no clear correlation with CDNC (lower panel), in four cases negative correlations are found: Chloride ($r^2 = 0.44$), organics ($r^2 = 0.33$), nitrate ($r^2 = 0.12$), and ammonium ($r^2 = 0.08$). An explanation might be that the variation from one cloud event to the other is too high to observe the effects of uptake from the gas-phase and in-cloud production.

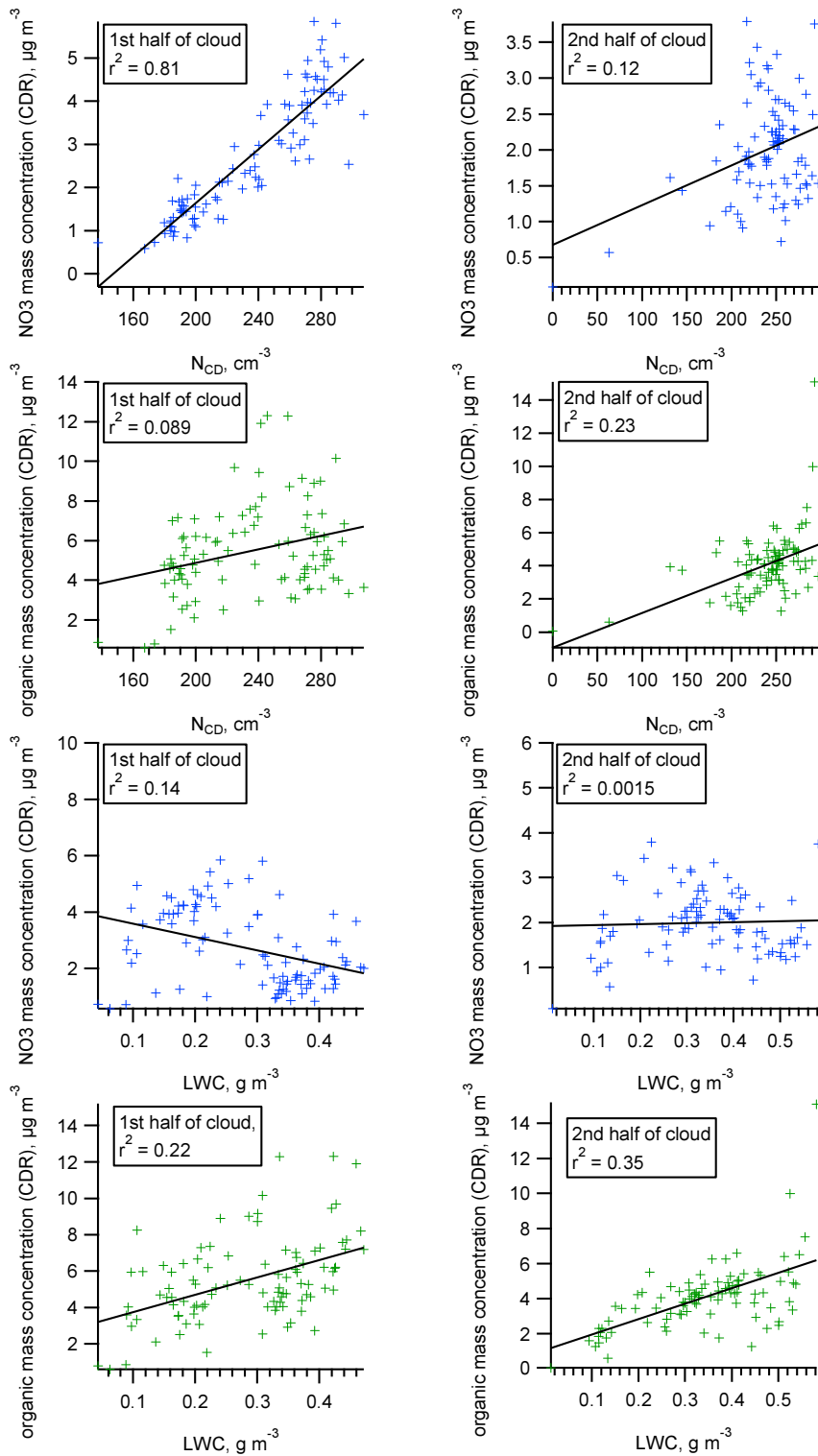
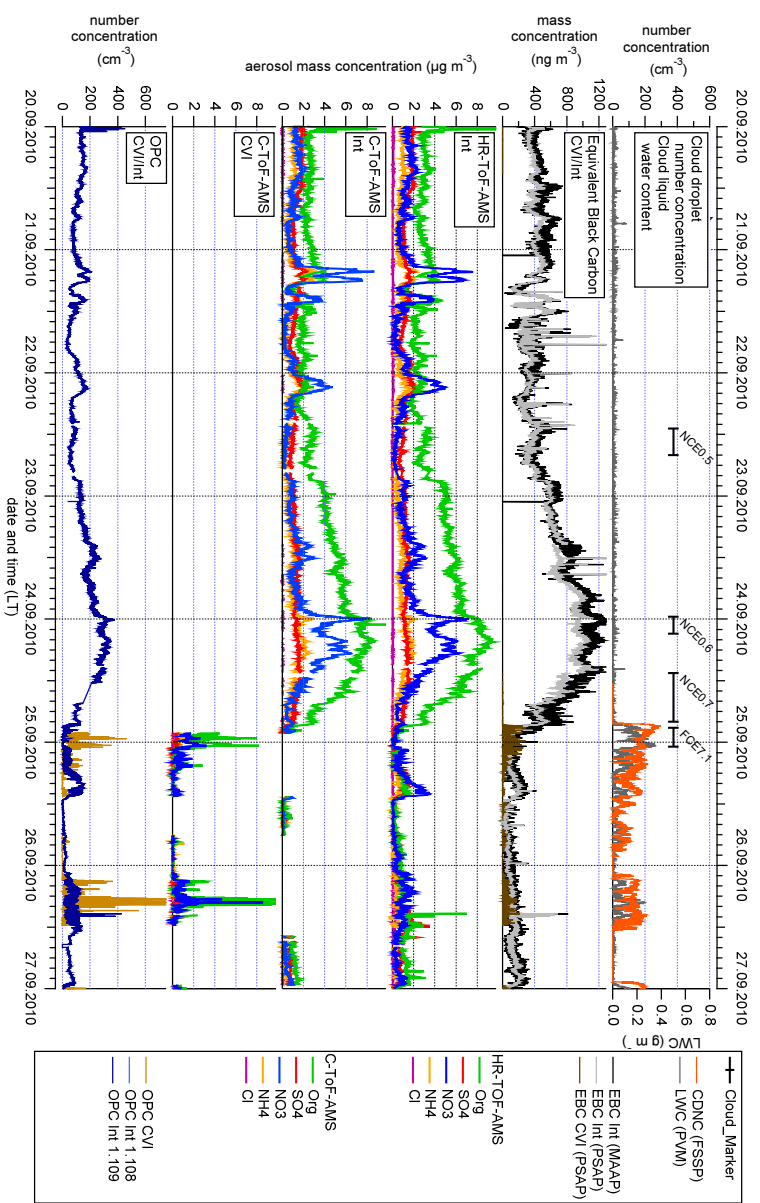
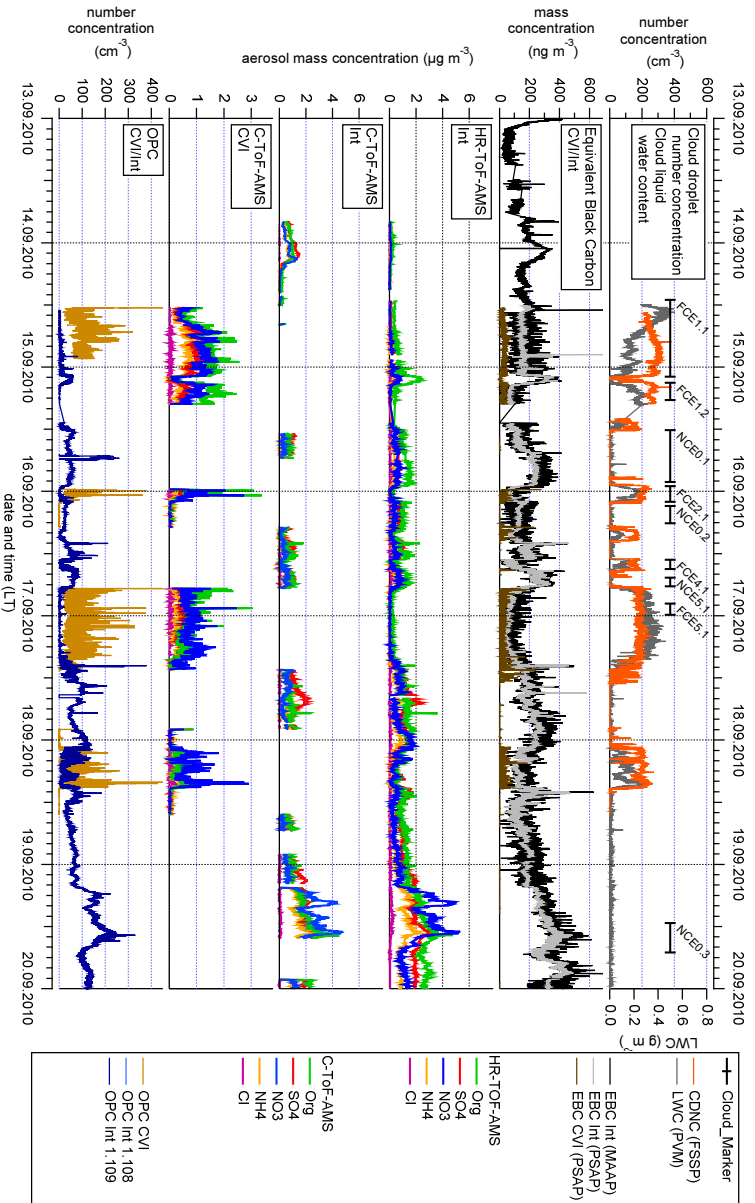
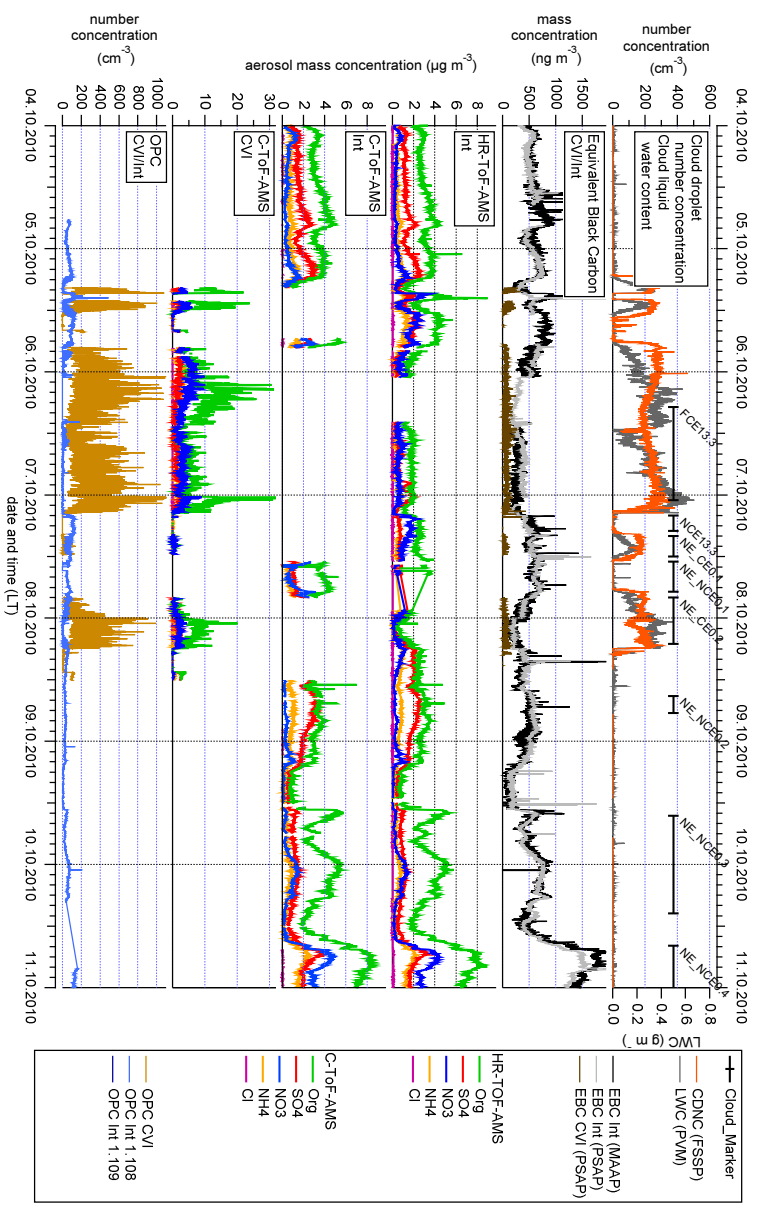
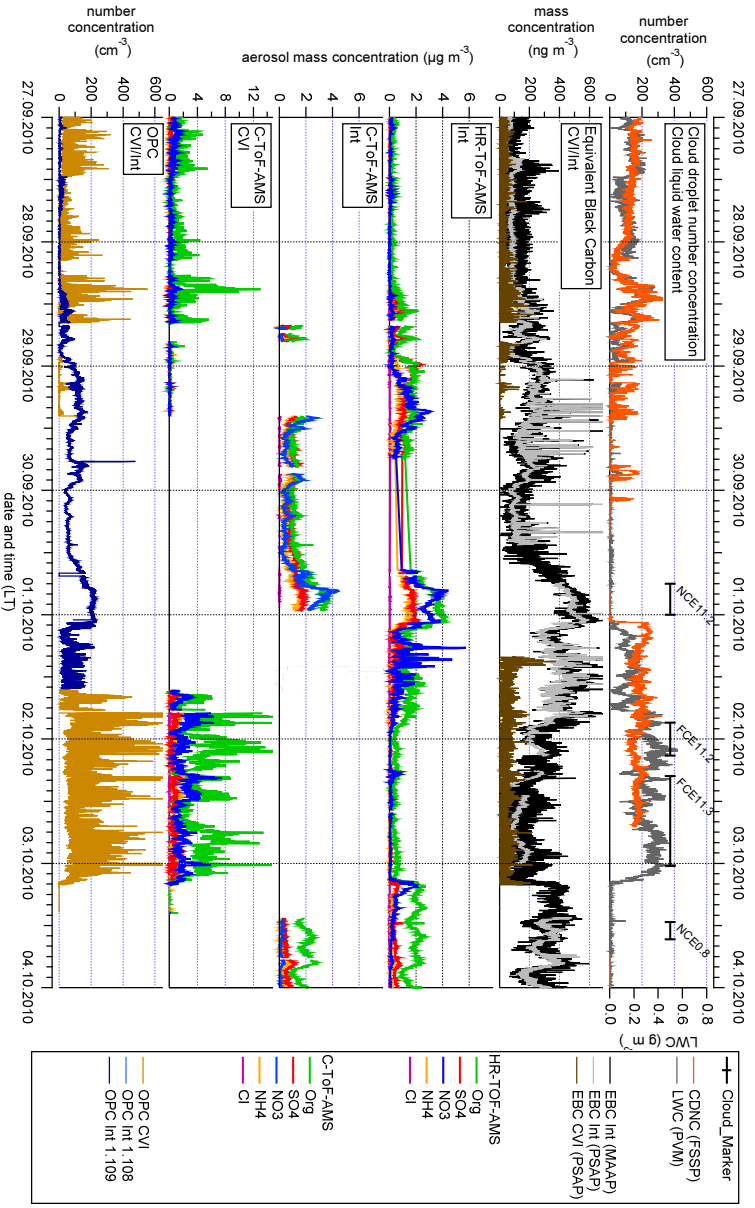


Figure S8: Mass concentrations of nitrate and organics in CDR vs CDNC and LWC during the cloud on 05 – 07 October 2010. Correlations were calculated separately for the first and second half of the cloud (first half: 05.10.2010 19:45 – 06.10.2010 11:55; second half: 06.10.2010 12:05 – 07.10.2010 03:35)





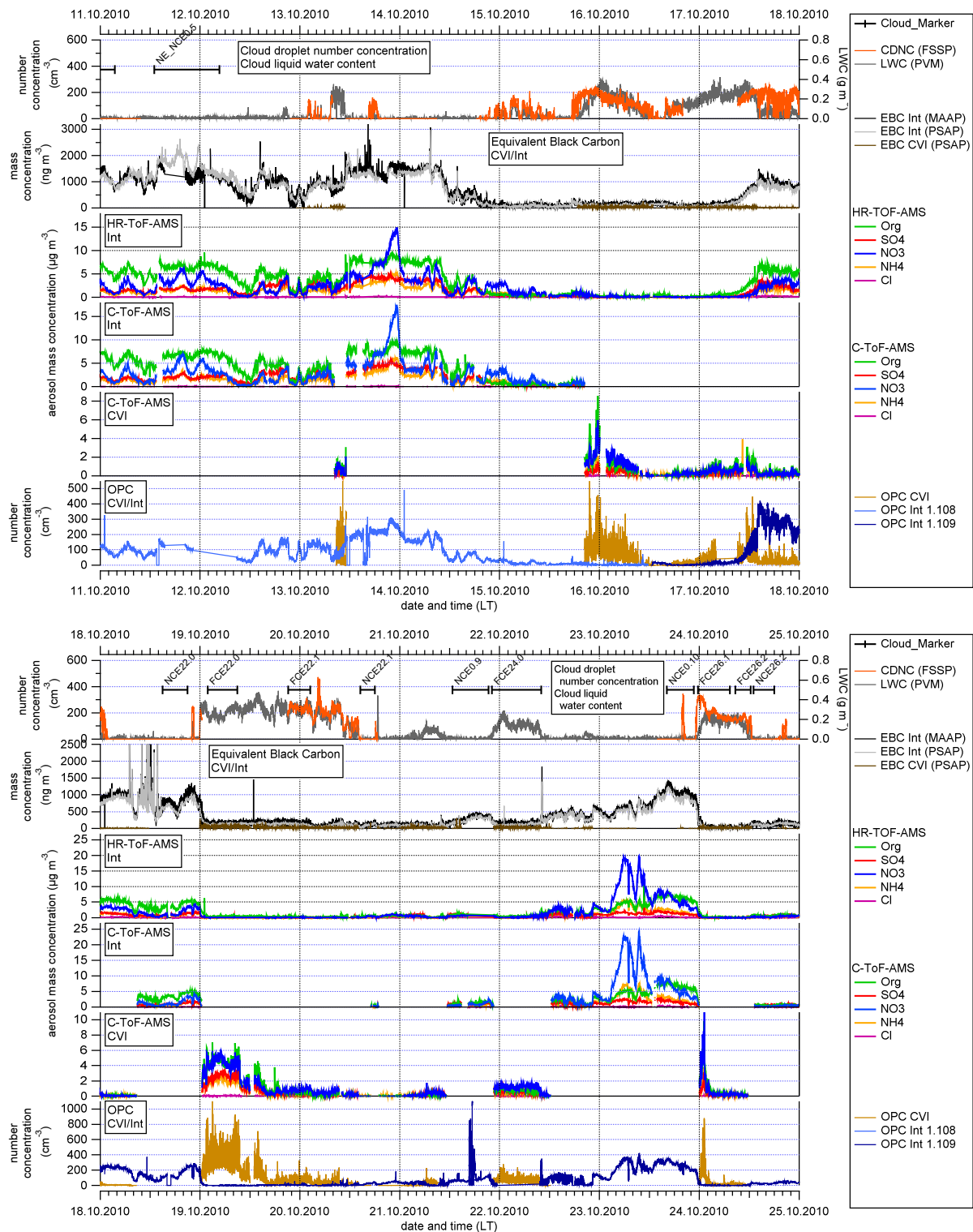


Figure S9: Times series of the measured parameters. In the upper panel the cloud events (FCE), non-cloud events ("NCE") and other reference periods as defined in Tilgner et al. (2014) are given. "Int" denotes the interstitial inlet.

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