

Figure S1 Influence of different pathways (SRR[%]pathway) averaged over the 5-month campaign.

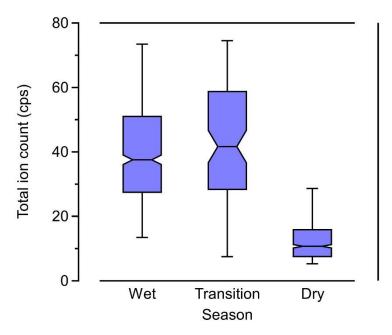


Figure S2 The TIC of negative ions measured by the APi-TOF in the wet, wet-to-dry transition, and dry seasons at the CHC. Data are in 1-hour time resolution.

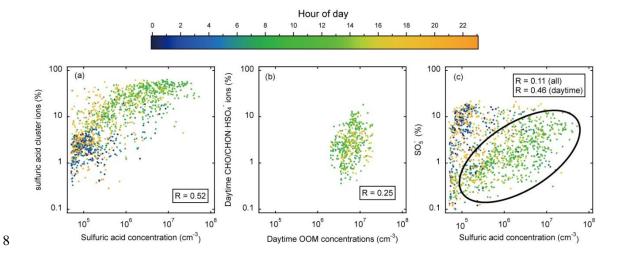


Figure S3 Relationships between (**a**) concentration of neutral  $H_2SO_4$  and signal fraction of the  $(H_2SO_4)_0$ .  ${}_3$ · $HSO_4$  ion group; (**b**) daytime concentration of OOM and signal fraction of CHO/CHON· $HSO_4$ ; and (**c**) concentration of  $H_2SO_4$  and signal fraction of  $SO_5$ . The black circle in panel (d) denotes the "daytime branch" of data points. The signal fractions of  $(H_2SO_4)_{0.3}$ · $HSO_4$  and  $SO_5$  ion groups were measured with the APi-TOF. Concentration of  $H_2SO_4$  was measured with the CI-APi-TOF.

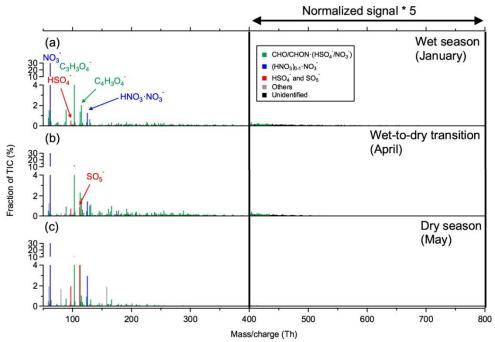


Figure S4 Mass spectra of negative ions at CHC averaged between 19:00 - 07:00 in (a) wet season (January), (b) wet-to-dry transition period (April), and (c) dry season (May). The normalized signal intensities from 400 Th to 800 Th are multiplied by a factor of 5 for better visualization.

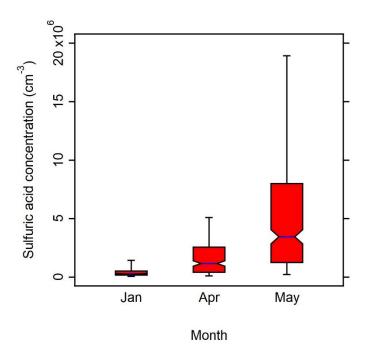


Figure S5 Daytime (07:00 – 19:00) neutral sulfuric acid concentrations measured with the CI-APi-TOF at the CHC during the study period when CI-APi-TOF data were available. Lower and upper limits of the error bars represent  $25^{th}$  and  $75^{th}$  percentiles, respectively.

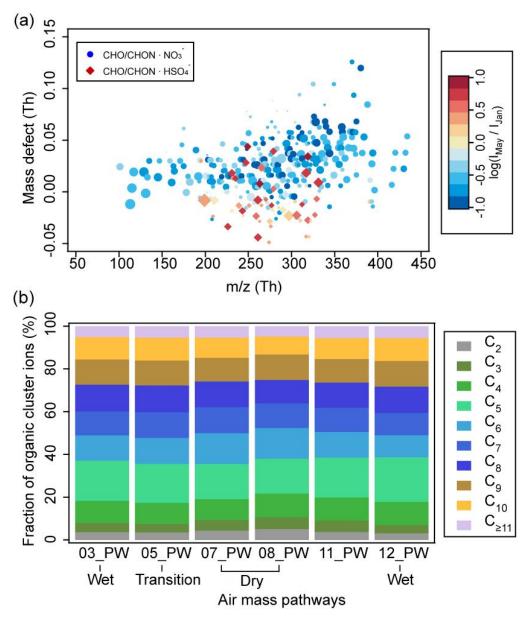
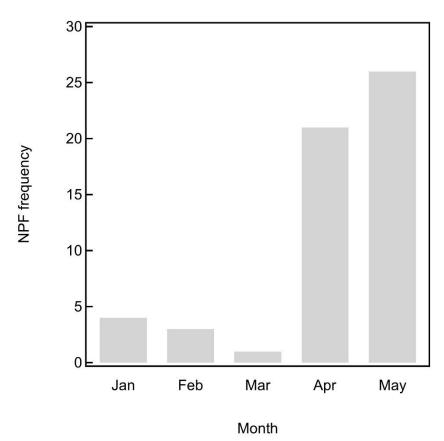


Figure S6 (a) Mass defect plot of organic clustr ions during daytime (07:00-19:00). The color code indicates ratios (in log scale) between median signals of each ion detected in May ( $I_{May}$ ) of dry season and January ( $I_{Jan}$ ) of wet season. The marker size is proportional to the log-transformed ion's median signals in May. (b) Fraction of organic cluster ions from different air mass pathways as a function of carbon atom numbers during daytime (07:00-19:00). Noted that MA-derived ions were not included in both panels.



Month
Figure S7 Frequency of NPF at the CHC from January to May 2018.

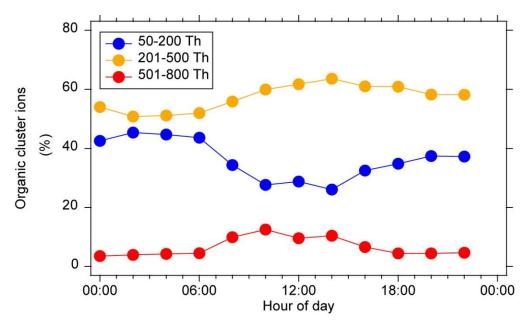


Figure S8 Variations of positive organic cluster ions in different mass ranges averaged during NPF events in February and March 2018.