



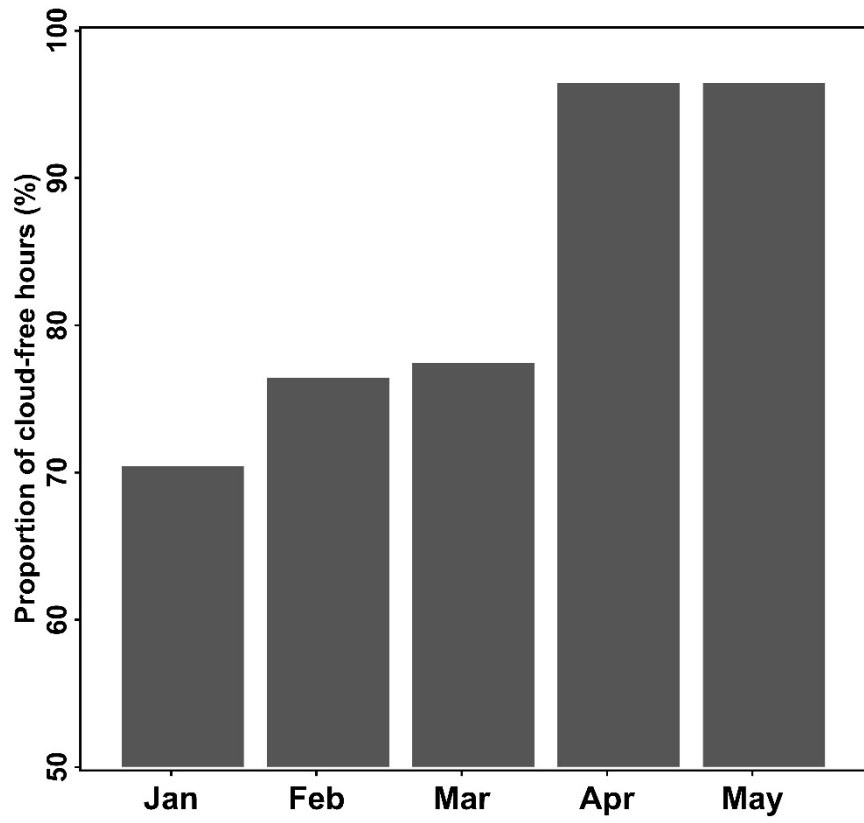
*Supplement of*

**Measurement report: Molecular-level investigation of atmospheric cluster ions at the tropical high-altitude research station Chacaltaya (5240 m a.s.l.) in the Bolivian Andes**

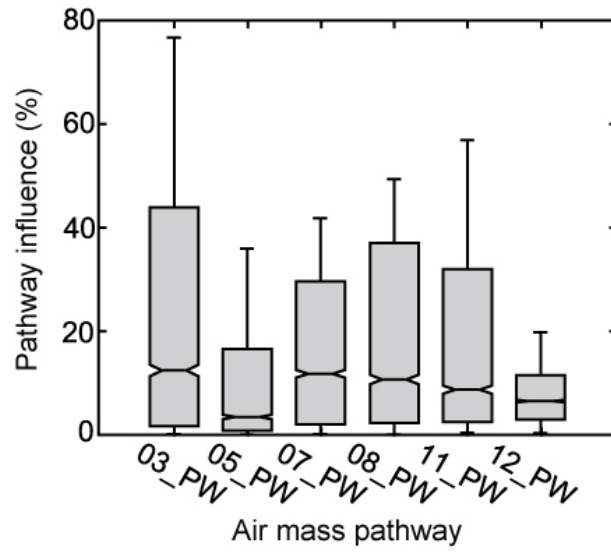
**Qiaozhi Zha et al.**

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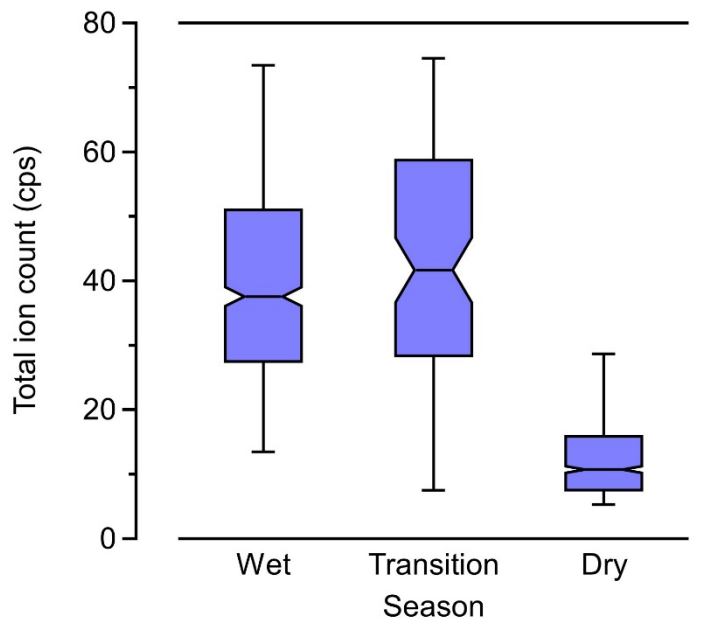
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2 Figure S1 Proportions of cloud-free hours to the total measurement time from January  
3 to May 2018.



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5 Figure S2 Influence of different pathways ( $SRR[\%]_{\text{pathway}}$ ) averaged over the 5-month  
6 campaign.

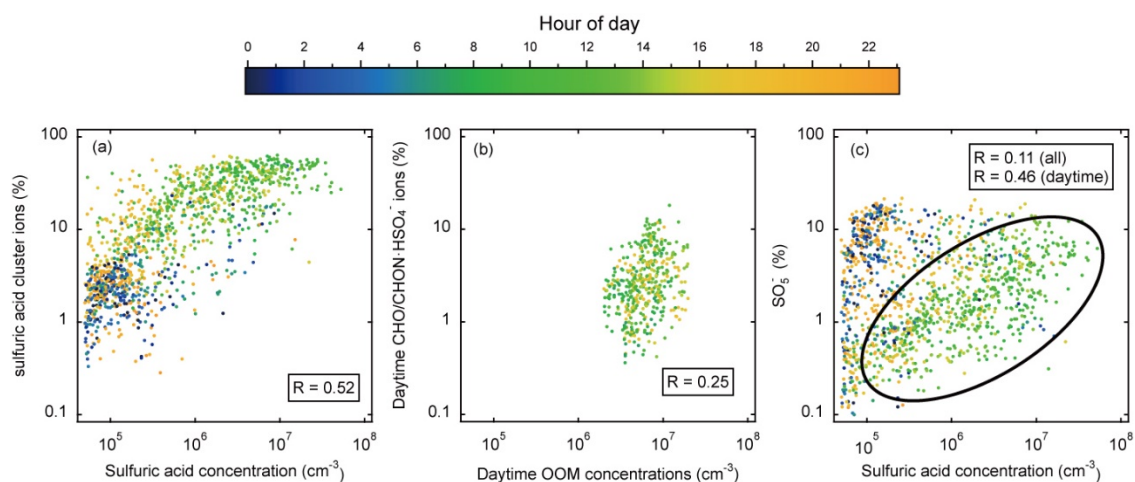
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9 Figure S3 The TIC of negative ions measured by the APi-TOF in wet, wet-to-dry  
10 transition, and dry seasons at CHC. Data are in 1-hour time resolution.

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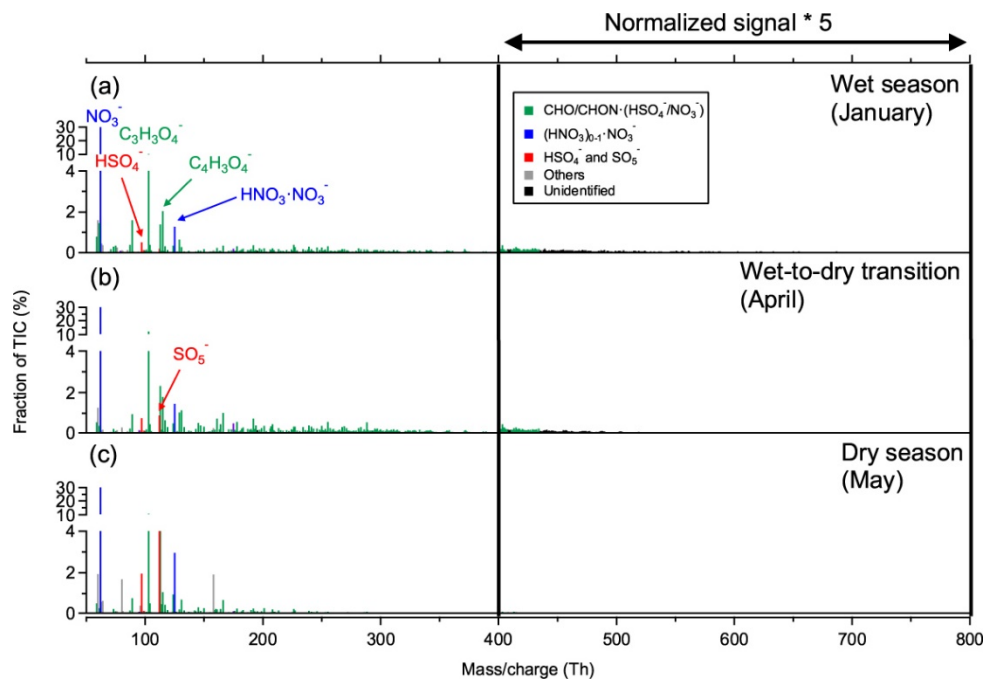


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13 Figure S4 Relationships between (a) concentration of neutral H<sub>2</sub>SO<sub>4</sub> and signal fraction  
 14 of the (H<sub>2</sub>SO<sub>4</sub>)<sub>0-3</sub>·HSO<sub>4</sub><sup>-</sup> ion group; (b) daytime concentration of OOM and signal  
 15 fraction of CHO/CHON·HSO<sub>4</sub><sup>-</sup>; and (c) concentration of H<sub>2</sub>SO<sub>4</sub> and signal fraction of  
 16 SO<sub>5</sub><sup>-</sup>. The black circle in panel (d) denotes the “daytime branch” of data points. The  
 17 signal fractions of (H<sub>2</sub>SO<sub>4</sub>)<sub>0-3</sub>·HSO<sub>4</sub><sup>-</sup> and SO<sub>5</sub><sup>-</sup> ion groups were measured with the APi-  
 18 TOF. Concentration of H<sub>2</sub>SO<sub>4</sub> was measured with the CI-APi-TOF.

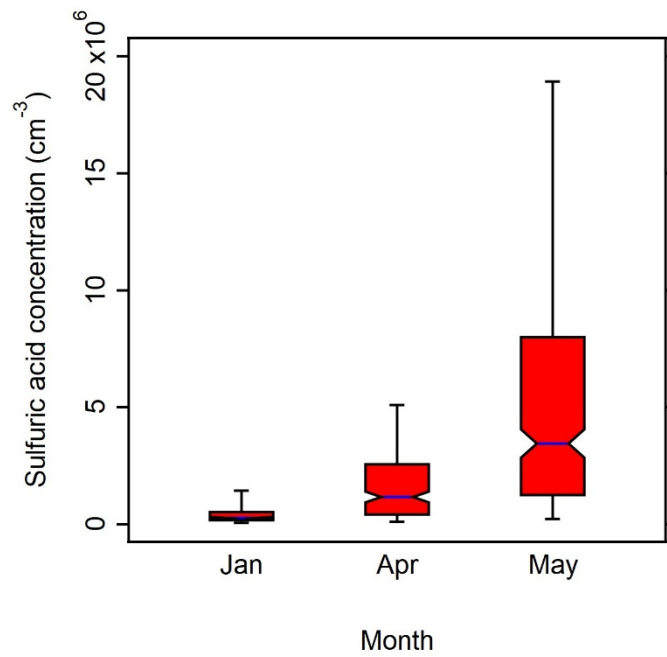
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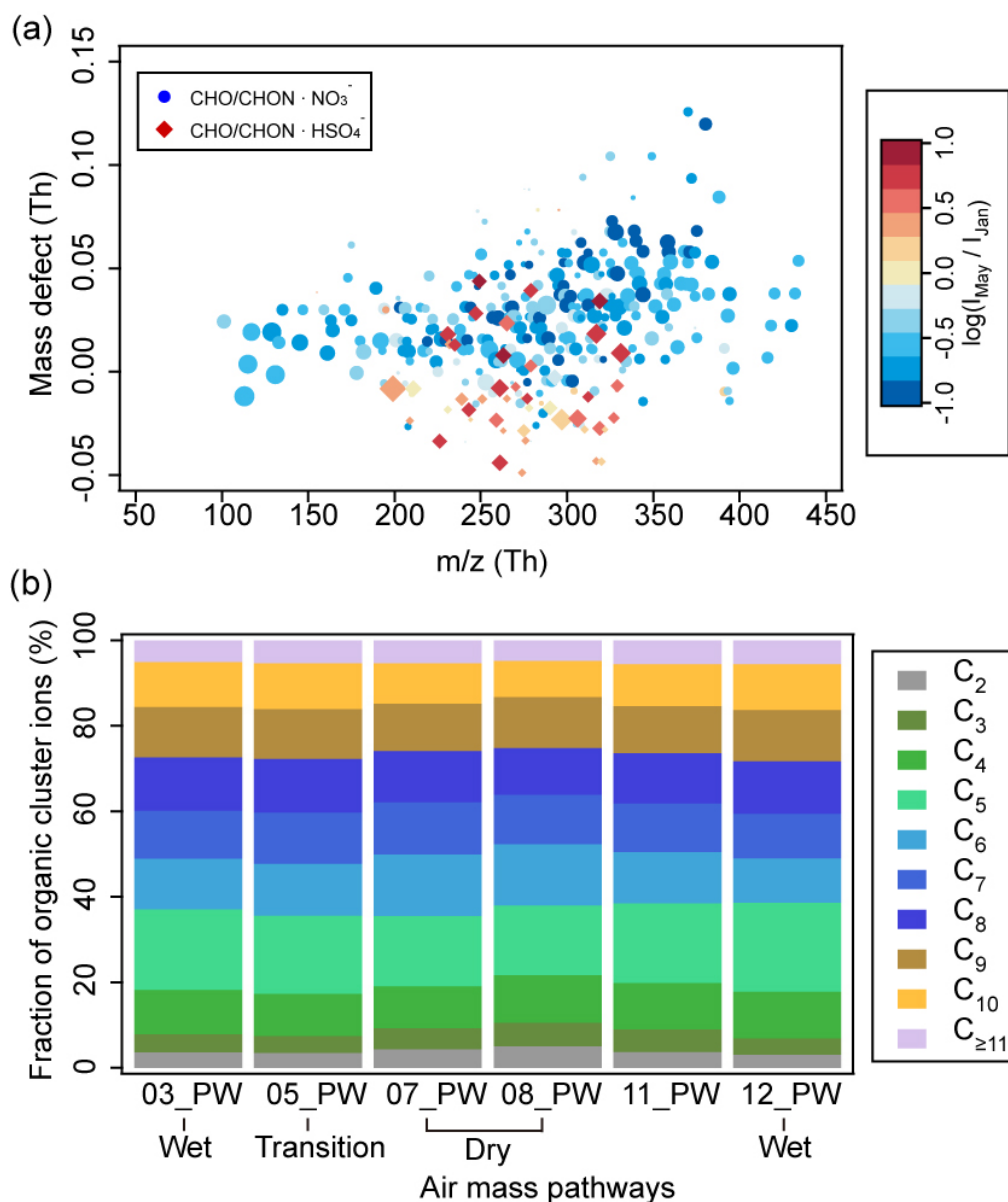
Figure S5 Mass spectra of negative ions at CHC averaged between 19:00 – 07:00 in **(a)** wet season (January), **(b)** wet-to-dry transition period (April), and **(c)** dry season (May). The normalized signal intensities from 400 Th to 800 Th are multiplied by a factor of 5 for better visualization.



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28 Figure S6 Daytime (07:00 – 19:00) neutral sulfuric acid concentrations measured with  
29 the CI-APi-TOF at the CHC during the study period when CI-APi-TOF data were  
30 available. Lower and upper limits of the error bars represent 25<sup>th</sup> and 75<sup>th</sup> percentiles,  
31 respectively.

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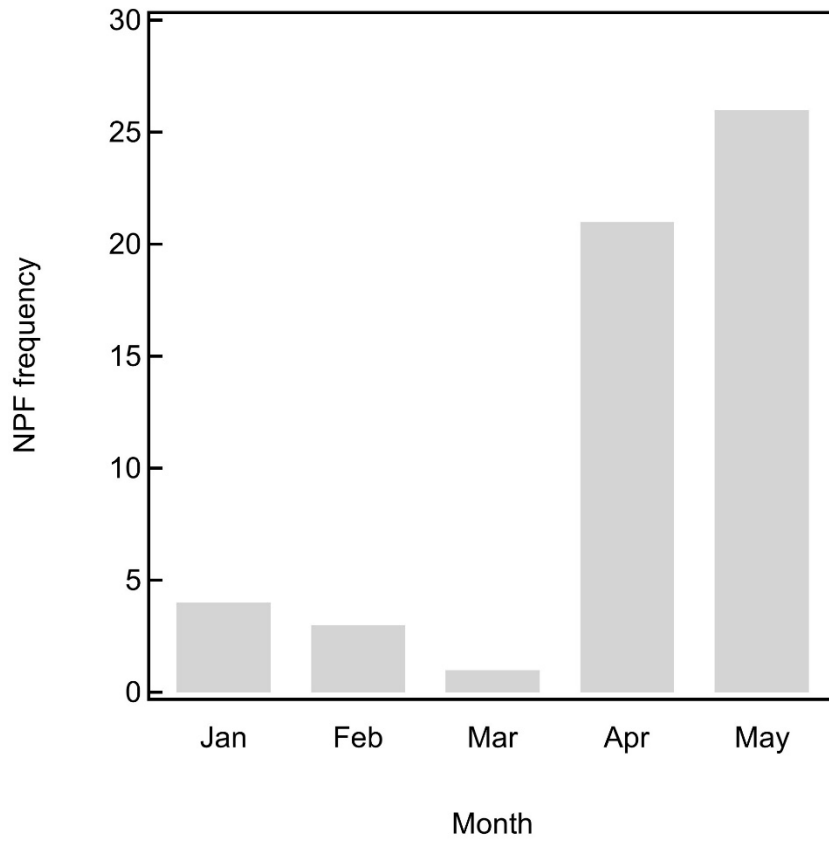
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35 Figure S7 (a) Mass defect plot of organic cluster ions during daytime (07:00-19:00).  
 36 The color code indicates ratios (in log scale) between median signals of each ion  
 37 detected in May ( $I_{\text{May}}$ ) of dry season and January ( $I_{\text{Jan}}$ ) of wet season. The marker size  
 38 is proportional to the log-transformed ion's median signals in May. (b) Fraction of  
 39 organic cluster ions from different air mass pathways as a function of carbon atom  
 40 numbers during daytime (07:00-19:00). Noted that MA-derived ions were not included  
 41 in both panels.

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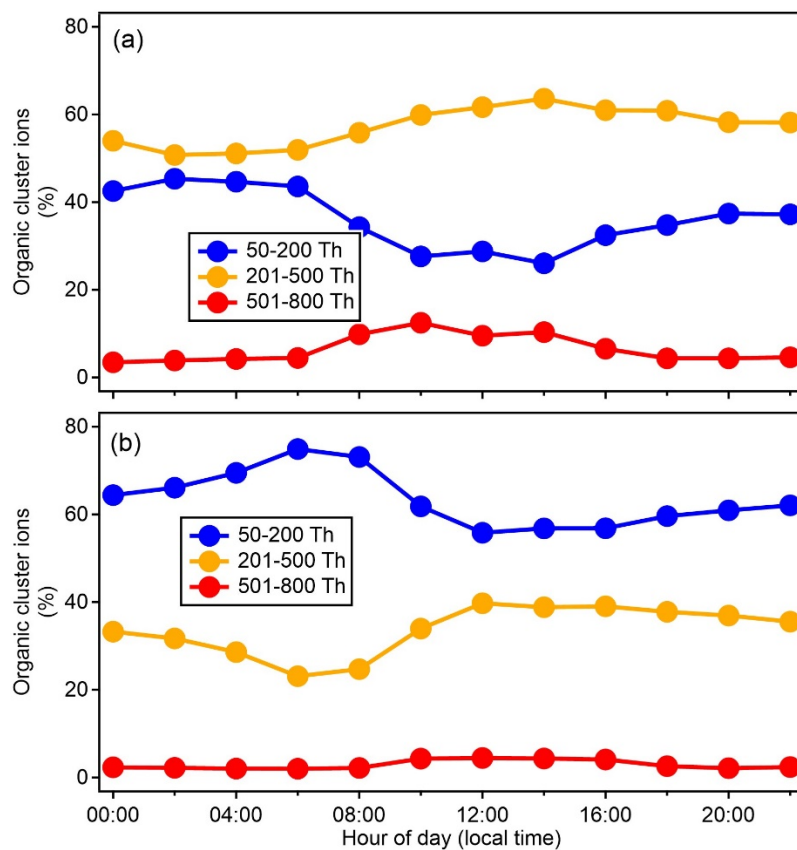




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45 Figure S8 Frequency of NPF events at CHC from January to May 2018.

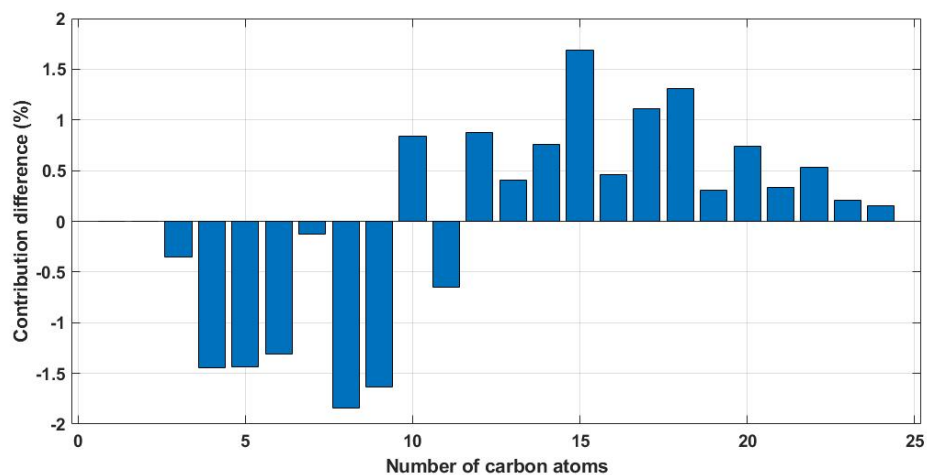
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48 Figure S9 Variations of positive organic cluster ions in different mass ranges averaged  
 49 during (a) NPF days and (b) non-NPF days in February and March 2018.

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 52 Figure S10 Difference in the identified positive organic cluster ions (median values)  
 53 observed before (06:00-08:00) and during (10:00-12:00) the NPF events in February  
 54 2018 (i.e., 18–19 February). Positive values refer to higher contributions during the  
 55 NPF events, and negative values higher contributions before the NPF events.

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