We appreciate the detailed comments and insightful suggestions from both reviewers. Our point-by-point responses are listed in green font. The corresponding modifications made to the revised manuscript are highlighted in light blue font. Please note that only references that are part of the replies to the comments are listed in the bibliography at the end of this document. References in copied text excerpts from the manuscript are not included in the bibliography. Line numbers refer to the updated track changes version of the manuscript.

**Reviewer 1 (RC1):**

This manuscript presents a large dataset of atmospheric cluster ions at a high-altitude station in the Sothern Hemisphere over a 5-month campaign, which is important. This work is one of the few existing studies reporting cluster ion composition at high altitudes, and probably the only one in the Southern Hemisphere. The chemical composition of the cluster ions was determined by using state-of-the-art instrumentation. The authors show interesting seasonal variations of the observed ions and attribute them to the properties of parent neutral molecules and different source origins by combing the results from the FLEXPART model. The potential link between the observed cluster ions and aerosol nucleation is also discussed. The manuscript is well-written and within the scope of ACP as a measurement report. I recommend it be accepted after the authors address several (minor) comments listed below:

***Reply***: We thank the reviewer for the positive and constructive suggestions.

1. While the authors attributed the variations of some cluster ions (e.g., NO3- and HSO4-) to the abundance and properties of their parent neutral molecules in a convincing way, it would be better to show the observed concentrations of, e.g., NOx and SO2.

***Reply***: Thanks for the suggestion. We agree with the reviewer that the connection between cluster ions and their parent neutrals can be better demonstrated by showing the concentrations of these neutral species (e.g., $NO_x$ and $SO_2$). Unfortunately, such measurements were unavailable due to instrumental issues during the study. Still, the dependence of cluster ions and their parent neutrals is evident, as we show in Fig. S4 and section 3.2.1 in the manuscript.

2. This reviewer understands that the seasonality of positively charged ions could not be determined because they were only measured in wet season. A significant fraction of the discussion in the manuscript is thus based on measurements of negatively charged cluster ions. However, as mentioned in line 554, the increase of large positive ions was found concurrently with NPF events. It would be better if the authors could specify the chemical composition of the NPF-related positive ions instead of the sum of the signals over a certain mass range.

***Reply***: As suggested by the reviewer, we have added a figure (as Fig. S10) showing the differences in the median contribution of the identified positive organic cluster ions

during two NPF events in February 2018. This provides more detailed information on the chemical composition of the NPF-related positive ions.

Figure S10 is added to the supplementary information and the following sentence is added to the revised manuscript (line 610):

"These organic cluster ions usually contained at least ten carbon atoms (Fig. S10)."

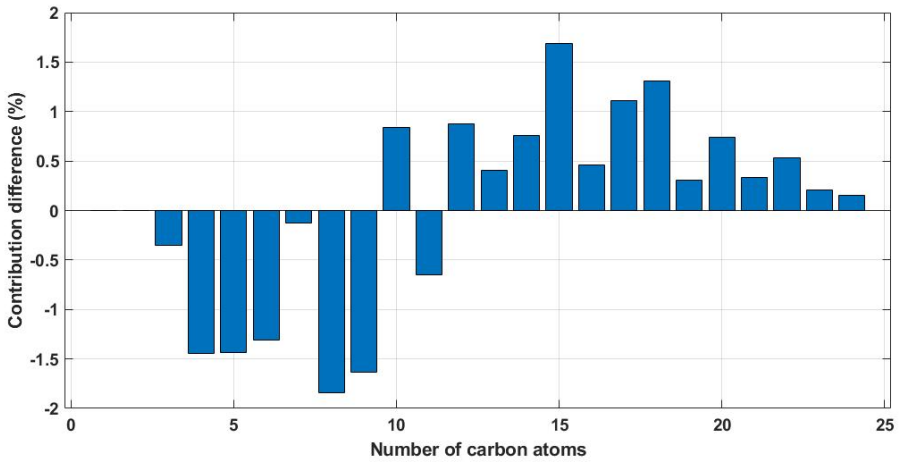


Figure S10 Difference in the identified positive organic cluster ions (median values) observed before (06:00-08:00) and during (10:00-12:00) the NPF events in February 2018 (i.e., 18–19 February). Positive values refer to higher contributions during the NPF events, and negative values higher contributions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

3. It is odd to see the fraction of SA-NH3 and SA cluster ions started increasing before the onset of nucleation (line 530), and an explanation for this may be needed. The reviewer suggests the authors make sure the aerosol data in Fig. 10 are synchronized with cluster ion data.

***Reply***: We thank the reviewer for the detailed comment and suggestion. After a thorough check of the data included in Fig. 10, we find that the size distribution of particles (Fig. 10a) was in UTC time, which is inconsistent with that of the cluster ions and sulfuric acid data (in local time, UTC + 4).

We apologize for this mistake and have updated Fig. 10 and revised the text in the revised manuscript (line 562):

"…always increased concurrently with the number concentration of small particles…"

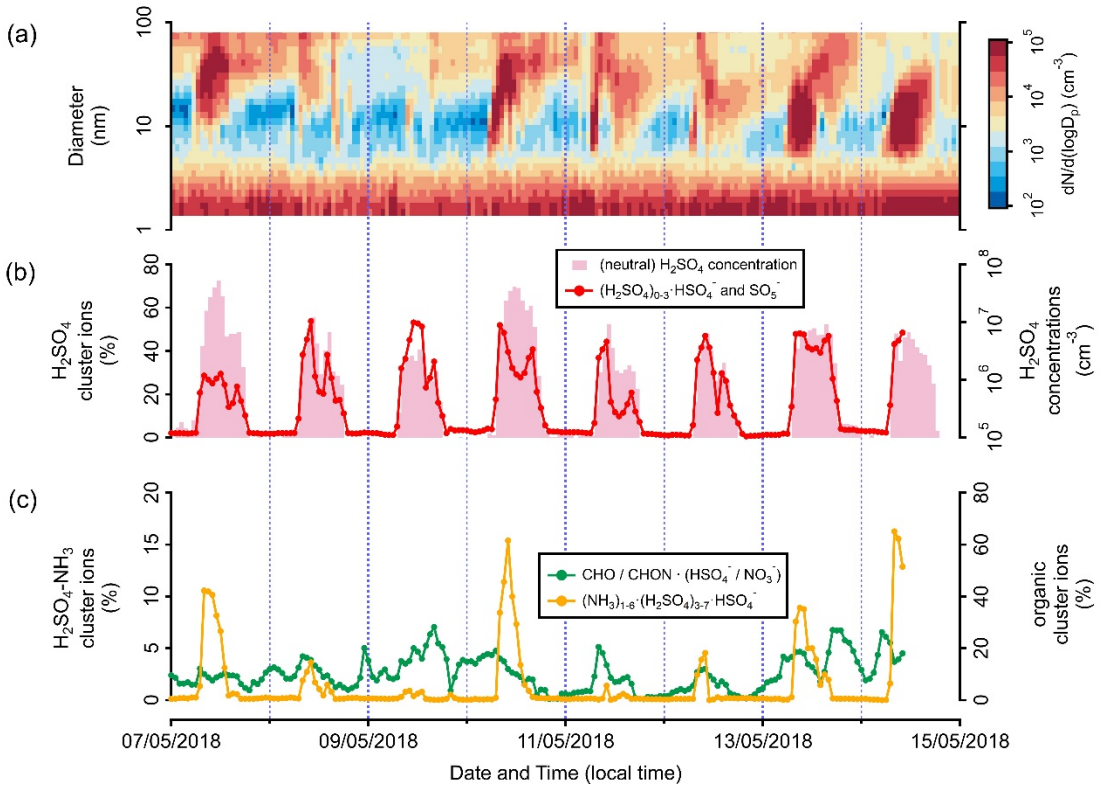


Figure 10 Time series of the **(a)** size distribution of aerosol particles (measured with NAIS and MPSS), **(b)** signal fraction of the $(H_2SO_4)_{0\text{-}3}{\cdot}HSO_4^-$ ion group and neutral $H_2SO_4$ concentration, and **(c)** signal fractions of the $(NH_3)_{1\text{-}6}{\cdot}(H_2SO_4)_{3\text{-}7}{\cdot}HSO_4^-$ and negative organic cluster ion groups, observed at CHC from 7 to 14 May 2018 when NPF occurred frequently.