Response to Editor Report

Thank you for your careful consideration of the referee comments. After thoroughly reviewing the response document and the revised manuscript and SI, I have determined that nearly all of the referee comments have been sufficiently considered. However, I feel that the manuscript would be improved by adding a few more details in response to reviewer 2 comment 1 ("The methodology regarding instrumentation is a little bit thin-on-the-ground. It would be nice to have more information about the equipment.") and 3 ("as your arguments hinge on the HOMs being monoterpene oxidation products, it would be nice if you showed them in more detail").

Response: Thanks for your suggestions, which help us to further improve the manuscript. The changes in the revised manuscript were highlighted in red color. Below is our point-by-point response to each comment.

Specifically, please consider adding information on the inlets (particularly important for something like H2SO4 and HOMs) as well as further information on how the MS was alternated between CI-APi-TOF and APi-TOF mode (line 144-145; e.g. how long in each mode?, how frequently was APi-TOF mode used?, etc.). Adding these details to the SI is ok.

Response: Thanks for the comments. The H_2SO_4 and HOMs were observed by CI-APi-TOF mass spectrometer. The sample air was drawn into CI-APi-TOF through a stainless-steel tube (100 cm long, 3/4 inch diameter) with a flow rate of 10 L min^{-1}. The sample flow was then surrounded by a clean sheath flow (25 L min^{-1}) containing nitrate reagent ions in a laminar flow reactor. The nitrate reagent ions in the sheath flow were generated by exposing air containing nitric acid to a photoionizer X-ray (Model L9491, Hamamatsu Inc.). To detect the natural cluster ions, the CI-APi-TOF mode was switched to APi-TOF mode by switching off the sheath flow and photoionizer X-ray and setting the ion voltage and drift voltage in laminar flow reactor to zero. On typical NPF (29 Apirl 2021) day, the mass spectrometer was alternated between CI-APi-TOF mode and APi-TOF mode at a frequency of once per hour from 10:00 to 21:00 (UTC+8), with each mode running 1 hour. We have added these details in the SI.

Regarding the HOMs being monoterpene oxidation products, while Fig. 4c is improved, it is still difficult to tell the intensity of the relevant HOMs in this type of plot. Additionally, as Fig. 4c is an APi-TOF measurement (or so it seems based on the caption), the HOMs distribution may be biased due to measuring naturally charged ions rather than neutral species. I encourage you to consider including something like an average MS (intensity vs m/z) from the CI-APi-TOF and coloring the sticks by the carbon number. This would more clearly show the intensity of the monoterpene oxidation products.

Response: Thanks for the comment. Indeed, the mass spectrum of natural ions clusters might be different with that of neutral molecules. According to your suggestion, we plotted the average mass spectrum of neutral molecules observed by CI-APi-TOF from 28 April to 30 April 2021. As shown in Fig. R1, the spectrum displayed the patterns of HOM monomer and dimer from monoterpene oxidation. The C5-10 HOMs dominated in terms of both species number and concentration. A certain concentration of HOM dimer was observed as well. It furtherly indicates the HOMs were mostly formed from monoterpene oxidation. We have added this result in the revised manuscript (Line 329-332 and Line 334) and Fig. R1 in the SI.
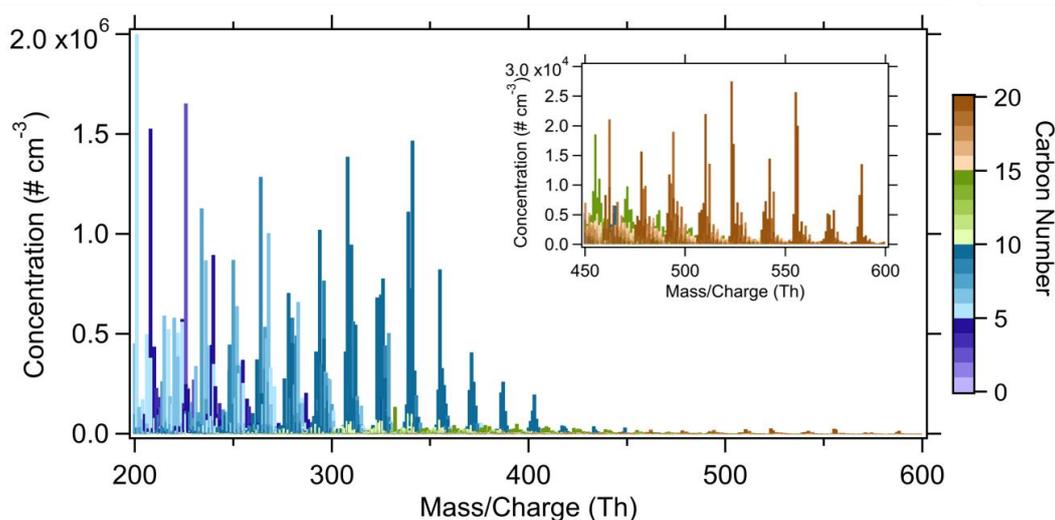

Figure R1. Mass spectrum of neutral molecules, colored by carbon number, observed by CI-APi-TOF during 28-30 April, 2021. Note that the top right subplot is zoomed-in view of m/z 450-600.

Line 331: "with the fraction of 32%." Please clarify what this is a fraction of. I presume it is of the HOMs ion intensity, but it is unclear.

Response: Thanks for the comment. The fraction is of the HOMs concentration. We have clarified it in the revised manuscript in Line 333.

As mentioned by the technical check, please ensure the color schemes are color blind friendly.

Response: Thanks for the comment. We have checked the Figures 1, 3, 4 using the Coblis and modified Fig. 4b.
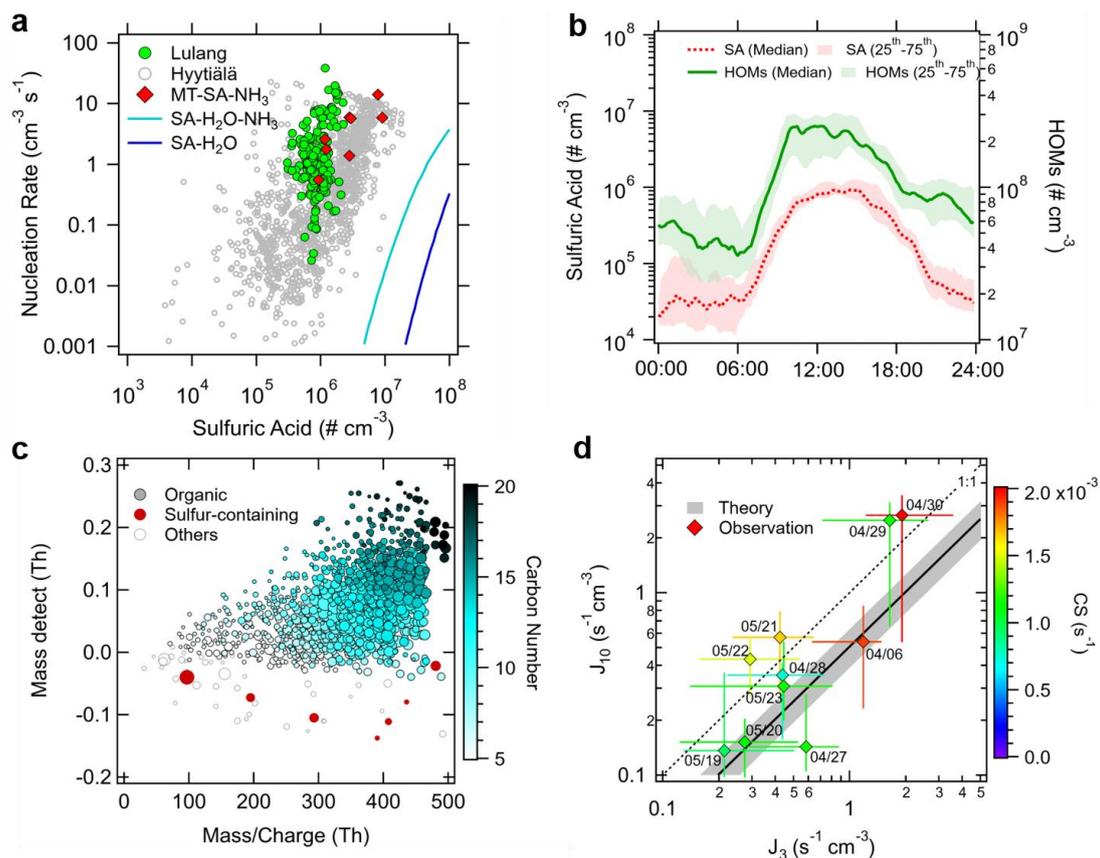
Figure R2. (a) Nucleation rates ($J_{1.7}$) as a function of H_2SO_4 concentration at ambient observations in Lulang (green circles), Hyytiälä (gray circles) (Sihto et al., 2006; Kulmala et al., 2013) and CLOUD experiments (red diamonds) (Lehtipalo et al., 2018). The cyan and blue lines denote ternary (H_2SO_4-NH_3-H_2O) nucleation and binary nucleation (H_2SO_4- H_2O), respectively, based on CLOUD data in Kürten et al. (2016). (b) Averaged diurnal variations of H_2SO_4 concentrations and HOMs concentration on NPF days in Lulang. The lines are the median values and shaded areas denote the 25th or 75th percentiles. (c) A mass defect plot illustrating the chemical composition of negative ion clusters at 12:00 (UTC+8) on 29 April. The size and color of symbol size correspond to the relative signal intensity on a logarithmic scale and carbon number, respectively. (d) Formation rate at 10 nm (J_{10}) versus formation rate at 3 nm (J_3) in Lulang. Diamonds are color-coded by condensation sink. Error bars present the 25th - 75th percentiles. The solid grey line shows the relationship between J_{10} and J_3 based on theory (Kulmala et al., 2012) and the uncertainties are shown by the shaded bands. Dash 1:1 line is shown for reference.