The following corrections are suggested to improve the readability and clarity of the manuscript, primarily in the abstract and introduction. Line numbers refer to manuscript showing tracked changes.

General comments:
Abbreviations such as “NPF” and “SOA” are used inconsistently throughout. It recommended to check that once these have been introduced they are used throughout the manuscript.

It is a little unclear whether the CIMS that is used with the TD-DMA is the same CIMS that is used for the gas phase (CI-APi-TOF). I think part of the confusion is that when discussed with the TD-DMA it is often referred to as the chemical ionization time-of-flight mass spectrometer (e.g., line 141) and not CI-APi-TOF. I think maybe this should just be CI-APi-TOF throughout.

The ratio, R, of isoprene to α-pinene is sometimes written as “R” and sometimes as “*R*”.

Specific comments:
The outstanding questions or hypotheses that this research seeks to address are a little unclear based on the abstract and the introduction. The sentences in the abstract are a bit disconnected, as are the paragraphs in the introduction. Some specific examples are given below as well as some specific suggestions for revision.

Abstract
Suggested revision of the abstract: “Biogenic organic precursors play an important role in atmospheric new particle formation (NPF). One of the major precursor species is α-pinene, which upon oxidation can form a suite of products covering a wide range of volatilities. Highly Oxidized Organic Molecules (HOMs) comprise a fraction of the oxidation products formed. While it is known that HOMs contribute to Secondary Organic Aerosol (SOA) formation, including NPF, they have not been well studied in newly formed particles due to their very low mass concentrations. Here we present gas- and particle-phase chemical composition data from experimental studies of α-pinene oxidation, including in the presence of isoprene, at temperatures (-50 °C and -25 °C) and relative humidities (20% and 60%) relevant in the upper free troposphere. The measurements took place at the CERN Cosmics Leaving Outdoor Droplets (CLOUD) chamber. The particle chemical composition was analyzed by a Thermal Desorption-Differential Mobility Analyzer (TD-DMA) coupled to a nitrate chemical ionization time-of-flight mass spectrometer (CIMS). CIMS was used for particle- and gas-phase measurements applying the same ionization and detection scheme.”

Line 61: change “are” to “were”: “…C_{15} compounds ($C_{15}H_{24}O_{5\text{-}10}$) were detected.”

No further changes to abstract are suggested.

Introduction
Lines 75-76, suggested revision: “…and are thus relevant for Secondary Organic Aerosol (SOA) formation, including New Particle Formation (NPF), due to gas-particle partitioning.

Lines 80-81 do not make sense as written. The sentence suggests that rapid growth occurs in α-pinene ozonolysis experiments across temperatures, but that there is also a reduction in the extent of autoxidation. It isn't really clear what is determined…the rapid growth? A decrease with temperature? An increase with temperature? Maybe it is being suggested that rapid growth is observed across temperatures because of compensating effects-higher autoxidation at higher T and increased partitioning at lower T due to decreased vapor pressures. This is clearer in the discussion of Simon, but is quite confusing in reference to Stolzenburg.

After the first paragraph introducing CCN and HOM, it is suggested that the authors then introduce isoprene and alpha-pinene and give an overview of what has been studied and where the gaps are. One suggestion is: "Isoprene (C_5H_8) has the highest global emission rate and many studies have demonstrated the importance of isoprene in terms of SOA formation. α-Pinene ($C_{10}H_{16}$), while less abundant, is one of the most commonly observed and prominent contributors to biogenic SOA. SOA formation has been well studied in isoprene and α-pinene systems. The role of HOMs in SOA formation and NPF also has been explored in α-pinene and α-pinene with isoprene systems. However, much less is known about the particle-phase composition of HOMs in these systems and the specific controls particle formation and growth rates, including as a function of temperature and the ratio of isoprene to α-pinene. "

A summary of the Kirkby, Stolzenburg, Kiendler-Scharr, etc. studies could then follow. The last paragraph could then be that "In order to better understand the roles of isoprene and temperature on HOM formation and associated rates of NPF, we present…."

Not sure that the sentence starting on line 106 "Additionally, some studies…" is needed. It does not add much to the specific discussion on HOMs, SOA, NPF, etc.