

The authors would like to thank the reviewer for the constructive comments. Our replies to the comments are given below, with the original comments in black, and our response in blue. We have revised the manuscript accordingly. All changes made to the manuscript have been marked with Track-Change tool in one of submitted files.

Anonymous Referee #1

This paper describes winter time NPF in the northeastern United states using the modeling predictions combined with ambient measurements of aerosol size distributions made at two sites and their contribution to CCN production. NPF is usually considered as a main source of CCN. And observations have shown that NPF usually takes place less frequently during the winter, in many locations over the world. So the results presented in this paper are interesting considering these factors. This is a well-written paper, easy to read.

We appreciate the referee's positive comments about this work.

Perhaps, the model used too high sulfuric acid concentrations to predict NPF winter?

We made long-term measurements of NPF and sulfuric acid in Ohio and our measurements show that winter time sulfuric acid does not exceed 3e6 per cubic centimeter (Erupe et al., 2010; Yu et al., 2014). And this paper claims that in order to have NPF, sulfuric acid higher than 3e6 per cubic centimeter is needed.

Related to this, at the same site in Ohio, we also found lower frequency of NPF during the winter than other seasons, very likely due to low sulfuric acid concentrations (Erupe et al., 2010; Kanawade et al., 2012).

This is a very good point. We agree that sulfuric acid concentrations ($[H_2SO_4]$) are critical for NPF. The model calculates $[H_2SO_4]$ through the balance between the photochemical production (SO_2+OH) and condensation. The model appears to generally capture the observed values and variations of SO_2 and solar radiations for the PSP site (Fig. 2b and Fig. 2c), both are important for the photochemical production of H_2SO_4 . Based on the model prediction, $[H_2SO_4]$ can reach $\sim 1E7/cm^3$ or higher and it is during these days that nucleation is significant (Figs. 2d and 2e). As emphasized in the paper, these nucleation events are necessary to explain the observed increase in CN10 (Fig. 2e).

It is true that the model predicted $[H_2SO_4]$ is higher than those observed (with CIMS) in Kent, Ohio during the winter (Erupe et al., 2010; Yu et al., 2014). The possible reasons for the difference remain to be investigated. One possible explanation is the well-recognized 1-2 orders-of-magnitude lower concentrations of sulfuric acid monomer measured with CIMS than the total-sulfate values measured with MARGA and the theoretical values calculated from the vapor pressure of sulfuric acid (Neitola et al., 2015).

We have added a discussion on this in the revised text:

“It should be noted that the model predicted $[H_2SO_4]$ is higher than those observed with a chemical ionization mass spectrometer (CIMS) during the winter in Kent, Ohio (Erupe et al., 2010; Yu et al., 2014), also located in the NEUS where wintertime nucleation was observed to occur on $\sim 17\%$ of days (Kanawade et al., 2012). The possible reasons for the difference of model-predicted and CIMS-observed $[H_2SO_4]$ remain to be investigated. One possible explanation is that sulfuric acid molecules are bonded with base molecules (e.g. ammonia and

amines), leading to the well-recognized 1–2 orders-of-magnitude lower concentrations of sulfuric acid monomers measured with CIMS than the total-sulfate values measured with MARGA and the theoretical values calculated from the vapor pressure of sulfuric acid (Neitola et al., 2015).”

Does the model consider temperature effects on nucleation and growth? NPF becomes more favorable at lower temperatures, as shown from laboratory studies (Duplissy et al., ; Yu et al., 2017; Tiszenkel et al., 2019). If the model includes this feature, then maybe this is due to lower temperatures?

Yes. Temperature is one of the most important parameters controlling NPF. Nucleation is favored at lower temperatures but other factors ($[H_2SO_4]$, $[NH_3]$, ionization rates, etc.) are also important. We have added a sentence emphasizing this point.

It would be nice to give some explanation why ternary ion nucleation (as opposed to neutral ternary nucleation) is important? What are the potential sources of ions in winter in the boundary layer?

The main reason is that charged clusters have a lower nucleation barrier and thus ternary ion nucleation is favored as opposed to neutral ternary nucleation. The details can be found in the reference (Yu et al., 2018) cited in the paper. The main sources of ions in winter in the boundary layer include galactic cosmic rays and radioactive materials from soils. We have added two sentences in the first paragraph of the Results session.

And is it possible to explain the growth rate from 3 nm to the CCN size with the sulfuric acid and ammonia? If not, what makes new particles grow so fast to become CCN?

As pointed out in the text (line 226 in the changes tracked version), equilibrium uptake of HNO_3 also contributes to particle growth in the winter.