Hirshorn et al., Responses to Reviewer 2

The authors of this paper would like to thank the reviewer for their insightful and constructive comments on the paper. We have carefully considered the feedback. Based on this feedback, we have made significant changes that have improved the work. Please note that references to line numbers below in the author responses correspond to the new line numbers in the updated manuscript.

The authors have color coded the responses to the reviewer as follows:

Blue: A response to the reviewer.
Black: Text that is in the originally submitted manuscript.
Red: Changes that were made to the manuscript and are reflected in the updated manuscript.

1. Line 86 – BVOCs can impact aerosol formation and growth.

Response: The fact that BVOCs impact aerosol growth as well as aerosol formation is now reflected on line 111 of the updated manuscript.

Addition: SPL is located above a mixed forest allowing for the emission of a variety of different biogenic volatile organic compounds (BVOCs) that can impact aerosol formation and growth (Amin et al., 2012).

2. Line 93-94 – Define Level 1, Level 2 and EBAS.

Response: A description highlighting the difference between level 1 and level 2 EBAS data is now included on lines 120 – 125. EBAS is the database of EMEP (European Monitoring and Evaluation Programme) EMEP is the co-operative program for monitoring and evaluation of the long-range transmission of air pollutants in Europe. EBAS is operated by the Norwegian Institute for Air Research. We credit the Norwegian Institute for Air Research on line 125.

Addition: ~~Level 1 and level 2~~ SMPS data from SPL are now available on the EBAS database (database of European Monitoring and Evaluation Programme) including level 1 data, which maintains 5 min time resolution while removing invalid values and calibrations, as well as level 2 data which presents hourly averages and quantifies atmospheric variability. Level 1 SMPS data is used in this study. The goal of EBAS data is to store long-term atmospheric science datasets and provide standards for quality assurance, thus rigorous standards for data quality are implemented to any data admitted to EBAS (Norwegian Institute for Air Research).

3. Line 127 – what is "ample"?

Response: In this case, ample means that the average number concentration below 25 nm for a given day falls above the 10^{th} percentile of all considered data. To better clarify what the word ample means, the sentence on lines 157 – 159 was edited.

Addition: For days ~~with ample~~ where the average particle concentration below 25 nm is above the 10th percentile of all data considered, the maximum of the Gaussians is calculated at each size bin.

4. Line 181 – ΔN8,Dmax rather then N

Response: We have corrected the value to $\Delta N_{8,Dmax}$ and the sentence on lines 221 - 223 now reflects this change.

Addition: Where ~~N~~ $\Delta N_{8,Dmax}$ is the change in the number concentration of particles across the size distribution from about 8 nm to the maximum diameter (about 340 nm)~~, and~~ during Δt which is the time difference from the defined start of an event to the defined end of an event.

5. Line 212 – It is unclear here why you bother with CCN for non-event days. Presumably, it is because you use this as a reference point for comparison of CCN impacted by events. The reason should be made clear here.

Response: The authors agree that the reasons for why events are compared to non-events needs to be highlighted. The goal with this comparison is to truly highlight the potential CCN enhancement of NPF events compared to days with no traces of NPF. By conducting this comparison on such a large dataset, we believe that this reference is suitable as it is specific to location, season, and diurnal cycles, . Multiple additions to the manuscript have been made below to better highlight the reason for comparisons.

Addition (Lines 257 – 261): We consider CCN concentrations during non-events to determine if NPF events result in an enhancement of CCN. ~~NPF and particle growth are largely influenced by sunlight, therefore,~~ Sunlight is generally necessary for NPF and growth; therefore, it is important to consider the variations in the seasonal diurnal cycle and obtain ~~four different~~ one unique value of CCN_{start} for each season that accurately represents the time that NPF impacts the site ~~for~~ during each season (Hallar et al., 2011).

Addition (Lines 279 – 289): To compare the impact NPF events have on CCN, CCN number concentrations directly measured are considered during the time period spanning from CCN_{start} to CCN_{end} during valid events and non-events. An average CCN number concentration for supersaturation levels between 0.2% and 0.4% is calculated for each individual time period. These values are then averaged each season separately between events and non-events. The goal is to determine whether CCN concentrations are enhanced by NPF events. During long-term studies, especially at clean, remote locations like SPL, directly comparing events and non-events will result in the relative enhancement of CCN due to events at a given location. By removing the subjectivity of selecting idealized cases, we provide a more robust methodology to evaluate long-term datasets. The methodology within this paper carefully considers similar timeframes within the diel pattern with and without NPF, to look at the relative change induced by NPF. By further comparing events to non-events through a seasonal lens, we ensure that days with similar meteorological conditions are compared. By further comparing events to non-events through a seasonal lens, we ensure that days with similar meteorological conditions are compared.

6. Line 213 – NPF and growth are impacted by sunlight as well as many other factors. Rather than "NPF and growth are largely impacted by sunlight, therefore…", I suggest something like "Sunlight is generally necessary for NPF and growth, and therefore…"

Response: The suggestion has been implemented on lines 256-258 of the edited manuscript.

Addition: Sunlight is generally necessary for NPF and growth; therefore, it is important to consider the variations in the seasonal diurnal cycle and obtain ~~four different~~ one unique value of CCN_{start} for each season that accurately represents the time that NPF impacts the site ~~for~~ during each season (Hallar et al., 2011).

7. Line 214-215 – Here you state that you "obtain four different values of CCNstart". I assume, but may be wrong, that you mean one for each season, but not four for each event, which is how it sounds. Clarify please.

Response: We have clarified that one value of CCN_{start}/CCN_{end} is determined for each season. This change is reflected on lines 256-258 as well as an additional clarification on lines 274-275.

Addition on 259-261: Sunlight is generally necessary for NPF and growth; therefore, it is important to consider the variations in the seasonal diurnal cycle and obtain ~~four different~~ one unique value of CCN_{start} for each season that accurately represents the time that NPF impacts the site ~~for~~ during each season (Hallar et al., 2011).

Addition on 277-278: Four different values of CCN_{end}, one for each season, are determined when finding CCN_{end} values for non-events.

8. Line 223-224 – It is written to sound like this is novel, yet surely it is obvious that you must allow a long enough time.

Response: We confirm that considering NPF overnight at our remote site is not a novel aspect of our work. However, the goal of this sentence is to show the reader that this aspect of NPF is not just considered by our method, but also why we decide to consider general particle growth patterns after the most intense period of NPF. This will hopefully clarify to the reader, and potential users of this method, that we have considered this aspect of NPF in the determination of our CCN consideration times.

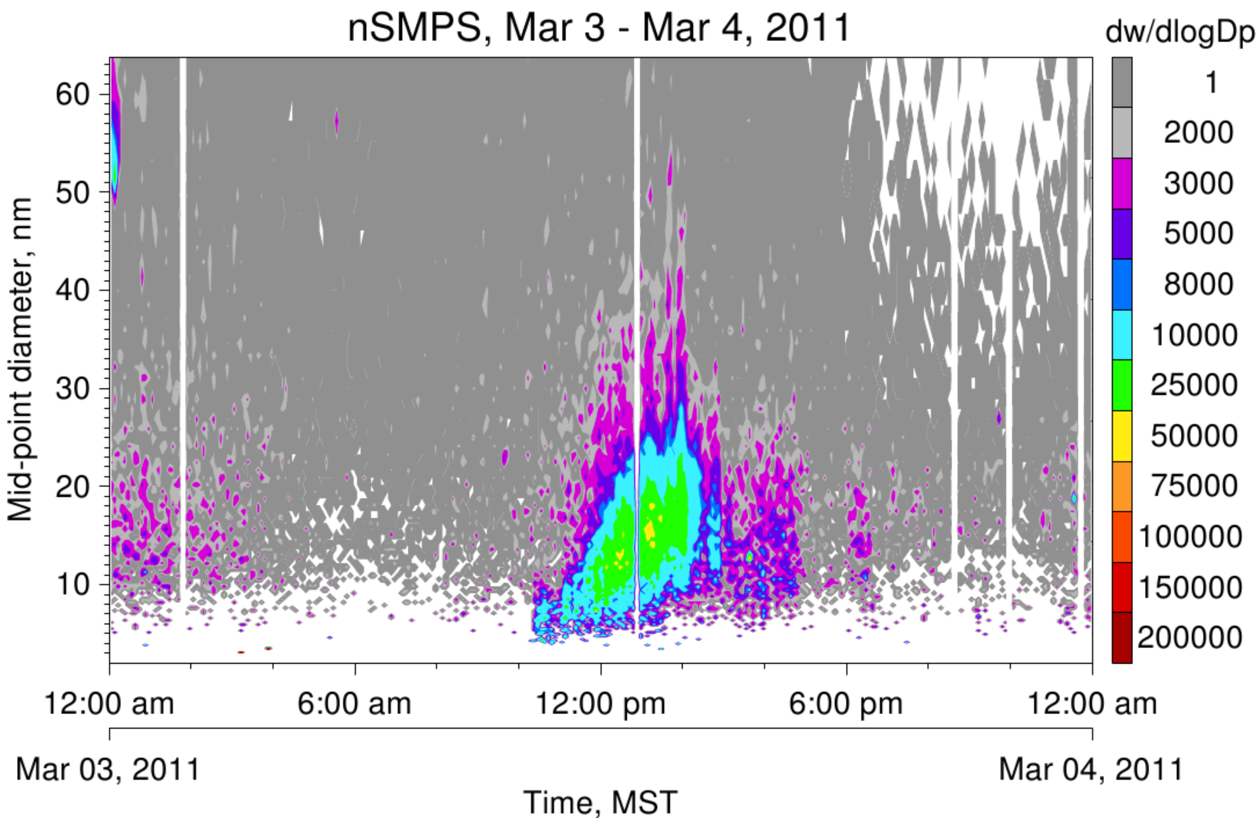
9. Lines 248- 250 - Is 53% significantly lower than 56% in this case?

Response: 53% is not significantly lower than 56% in this case. To emphasize this point, the sentence on lines 305-306 was edited.

Addition: Spring (53%) and winter (41%) display similar but slightly lower event frequencies than the summer and fall at SPL.

10. Lines 260-263 - What is the method of production of particles on non-event days, and are they truly new particles or just the result of a meteorological change, such as a developing boundary layer (since you are at a mountain site)?

Response: This is a great point and something that needs to be addressed to confirm that NPF days at SPL truly do represent a contribution of new particles compared to non-event days. In Hallar et al., 2016, a nano-SMPS was used to show that NPF events are accompanied with a burst of particles that is observed as low as 5 nm. Below is an example of a size distribution from the nano-SMPS:



By observing these particles at such low sizes, we have added confidence that observed particles during event days are newly formed and thus NPF, not solely transported through a meteorological change. Data from the nano-SMPS is only available for 1 year in our data collection period which is why this analysis is not included in this long term study. However, to emphasize that there has been previous work to highlighting that these particles are indeed due to NPF, a sentence on lines 324-325 was added:

Addition: Previous work at SPL has shown that during NPF events, particles as low as 5 nm are observed alongside events demonstrating that particles observed during NPF originate from nucleation (Hallar et al., 2016).

11. Line 265 – I suggest changing "produce" to "indicate".

Response: This change has been made on Line 322.

Addition: Above 82 nm, days with NPF events do not ~~produce~~ indicate more particles than non-events, which suggests any enhancements in CCN due to NPF events are likely due to particles below 82 nm.

12. Lines 266-267 - I'm having trouble understanding the apparent use of non-events as a reference. Why is it necessary to use non-events this way. Are you assuming that days with events would be the same as non-event days, IF there was no event? Presumably, one of the triggers for the event could be that the aerosol immediately preceding the event was particularly low in number and size (i.e., low CS). Would the non-event reference be appropriate in that case? Also, it is possible that the aerosol sampled on non-event days may well have been influenced by NPF at some time in its history. Would you clarify your reasons behind this, please?

Response: The justification for putting the section that details Figure 4 in the paper is to highlight that more particles below 82 nm in diameter are produced during events which emphasizes a quantifiable difference between events and non-events at SPL that could potentially impact CCN.

Given that there is a 15-year dataset and multiple event and non-event days for each season, we believe that comparing type 1a and type 1b NPF events to non-events is the best way to compare the contribution of CCN from days with an NPF event compared to days where this phenomenon is completely absent. At a mountaintop observatory, there is no such thing as a perfect NPF event or a perfect non-event. However, the purpose of having such a long dataset is that days with an anomaly will be less prominent in the comparison as they are averaged out by the more characteristic NPF events and non-events. This ensures that the highest number of days possible are included in this comparison providing the best overview of the relationship between NPF events and non-events at SPL.

One aspect of the code that helps ensure a quality comparison is the undefined classification category. The undefined classification category includes days where there is evidence of elevated aerosol concentrations but do not exhibit the growth of an event. This category helps to capture days without NPF events that have higher aerosol concentrations ensuring that the non-event category represents clean conditions at SPL absent of any burst like behavior that could skew the comparison.

Furthermore, as shown in Hallar et al., (2011) and here, we do not see a significant difference between the CS before NPF events and on days without NPF events throughout the entire dataset. Thus, in this clean remote region, it appears the CS does not solely drive the occurrence of an NPF event.

13. Line 283 - Here, the summer result again suggests the "non-event" may not be a suitable reference.

Response: This response builds on the response to the above comment. The summer result showing more CCN during non-event days compared to event days was not what we were expecting to find. However, this comparison is an important result that highlights why we conduct the comparison from a seasonal lens. As detailed in the paper, the summertime displays different conditions (wildfires, higher temperatures leading to BL fluctuations, organics) than other seasons. What this shows us is that while days with NPF events produce CCN, they are not a significant contributor due to other factors that can influence days without an NPF event. The seasonal comparison helps us conduct an accurate comparison while identifying the seasons where this relationship may not be true as well as the factors that influence this relationship.

14. Line 303 – What about condensable organics? Do higher temperatures somehow inhibit growth by organics? How would your CCN number concentrations react if organics played a major role in growth of the newly formed particles, compared with sulphate? What do you think are the main precursors leading to particles growth at SPL?

Response: We found it best to format the response by responding specifically to one question at a time.

What about condensable organics? Do higher temperatures somehow inhibit growth by organics?

The potential role of organics was left out of the paper because the relationship between temperature and particle growth by organics is complex and non-linear. This relationship is highly dependent on the organic species that is observed and was explored by Stolzenburg et al., 2018 (DOI: https://doi.org/10.1073/pnas.1807604115) finding that higher temperatures can lead to increased reaction rates and concentrations of highly oxidized organic molecules. However, lower temperatures can help decrease volatility resulting in less oxidized species becoming able to condense. Thus, there is not only a reliance on temperature, but also a reliance on the degree a molecule is oxidized. While obtaining measurements of organics that can detail the degree of oxidation would be highly beneficial, it is outside of the scope of this work since we do not have that data available.

How would your CCN number concentrations react if organics played a major role in growth of the newly-formed particles, compared with sulphate?

The authors believe that this is a question that is beyond the scope of this work. To answer this question, we would need access to the specific types of organics that are observed at SPL during each season, SO_2 measurements to compare the general organic:sulfate ratio, and aerosol composition measurements. This is a fantastic question that could be a great idea for future work but we do not currently have the measurements to conduct a clear comparison.

What do you think are the main precursors leading to particles growth at SPL?

We believe that the main precursor leading to particle growth at SPL is the presence of H_2SO_4 that is formed due to SO_2 emissions from the powerplants upwind. Lower temperatures that allow for nucleation to occur are another important factor. UV radiation and westerly winds (location of powerplants) have also been proven to lead to more favorable conditions of NPF. The role of organics has yet to be investigated in depth. The impact organics may have on NPF at SPL is highly dependent on the type of organics observed and will require further work to make a conclusion.

15. Lines 310-333 - You have more bursts in summer and fall that suggests H2SO4 is being produced. Are you suggesting that it ends up condensing on existing larger particles, thereby improving their ability to act as CCN? In the first paragraph here, you say that a reduced CS is important for your SPL observations of NPF, and that is shown in Table 2. However, then you lead off the second paragraph stating that one phenomenon is influencing NPF and CCN on event days at SPL in the summer: temperature. I feel that this is misleading without a detailed analysis of the many things that might affect NPF: including, temperature, CS, SO2 concentrations, irradiance, available condensable species. In relatively clean environments with low concentrations of precursors (e.g., Arctic), a low CS can be to be a trigger for NPF. Higher concentrations of SO2 superimposed on regions with a low CS will result in higher number concentrations of NPF. Higher SO2 concentrations in regions with a higher CS will tend to have lower NPF. In other words, the CS is important, along with other factors. Unless you can present more evidence or a stronger argument that temperature is the primary factor controlling NPF here, I think the focus on one factor may be misleading and the discussion should be made a little more objective.

Response: The authors agree that it is important to ensure that the discussion reflects the work done in the paper and is not overly objective or accidentally misleading. To address this concern, language in the discussion has been added and/or changed to be less objective and reflect the scope of the work. Lines 345-397

Additions: NPF significantly enhances CCN concentrations in the spring and winter, the two seasons with the highest frequency of type 1a and type 1b events. Previous work at SPL indicates that the increased prevalence of anthropogenic H_2SO_4 precursors and cooler temperatures are two potential reasons that can lead to conditions that are conducive to NPF during the spring and winter seasons (Hallar et al., 2016; Yu and Hallar, 2014). While previous laboratory studies suggest that multiple gases including ammonia, amines, and organic compounds all influence NPF, H_2SO_4 is important for initiating particle nucleation due to its low volatility under atmospheric relevant conditions (Yu et al., 2015; Sipila et al., 2010). SO_2, which is a precursor of H_2SO_4, is emitted from coal-fired powerplants upwind of SPL allowing for the transport of SO_2 which has been previously observed at SPL and can help explain the high frequency of NPF events (Hallar et al., 2016). In addition to H_2SO_4, lower temperatures are another important factor that can aid the enhancement of ~~enhances~~ particle nucleation by lowering the thermodynamic energy barrier required for nucleation to occur (Yu, 2010; Bianchi et al., 2016; Duplissy et al., 2016; Lee et al., 2019). The combination of prevalent H_2SO_4 precursors and lower temperatures are two possible factors that can allow for the occurrence of persistent NPF on a regional scale during the spring and winter (Yu and Hallar, 2014). These results from modeling work suggest the significant enhancement of CCN due to NPF events during the winter and spring at SPL may be applicable on a regional scale in remote regions of North America downwind of power plants providing insight into the processes that drive CCN formation.

NPF does not significantly enhance CCN concentrations in the summer and fall seasons, compared to the spring and winter seasons (Figure 5). ~~, likely due to~~ One factor that could help explain this phenomenon are higher temperatures observed in the summer and fall compared to the spring and the winter. Higher temperatures in the summer and the fall, the seasons where NPF is not significant for forming CCN, can be ~~are a~~ barrier to nucleation since higher temperatures lead to lower supersaturation ratios of H_2SO_4 (Yu et al., 2015). In addition to higher temperatures, previous work shows that SO_2 concentrations at SPL are slightly lower in the summer and fall than in the spring and winter, suggesting that H_2SO_4 could be less likely to form due to the combination of higher temperatures and lower available SO_2 (Hallar et al., 2016; Yu et al., 2015). SO_2 is not available for the entirety of the dataset, hindering the direct connection between H_2SO_4 precursors to the occurrence of NPF at SPL.

The CS and environmental conditions are two additional factors that can potentially explain the presence of higher aerosol concentrations during the summer and fall, despite ~~coupled with~~ the lack of a CCN enhancement due to NPF. The CS is a parameter that indicates how fast aerosols will condense onto pre-existing particles while also indicating how many pre-existing particles are present (Kulmala et al., 2001; Pirjola et al., 1999). Table 2 shows that CS values are highest in the summer, followed by the fall at SPL, indicating there is more pre-existing aerosol in the summer and fall than in the spring and winter. Data from the Whistler Aerosol and Cloud study, which also takes place in a montane setting in western North America, also finds that particles are more likely to grow to CCN relevant sizes when the CS is lower since there are fewer particles to react with condensable gases, a trend that is also observed in this work (Pierce et al., 2012). The role the CS has on NPF is highly dependent on the conditions of a given site which is why it is important to report CS values. Environmental conditions in the Intermountain U.S., such as wildfires, are another factor that could help explain the higher CCN concentrations present in the summer and the fall during both events and non-events since aged smoke has been observed to enhance CCN concentrations at sizes above 80 nm in the western US (Twohy et al., 2021). With wildfires becoming more frequent in the western US, CCN from wildfire emissions is expected to be a contributor to total CCN during the summer and fall months at SPL (Hallar et al., 2017). More work is needed to better understand the role that the CS and wildfires play on CCN at SPL during the summer and the fall.

The lack of a significant CCN enhancement by NPF at SPL during the summer suggests that one potential phenomenon influencing NPF, and eventually CCN concentrations, is that lower temperatures are lowering the energy barrier required for H_2SO_4 formation in the winter and spring (Yu et al., 2015). This suggests that an anthropogenic source of SO_2, similar to the powerplants upwind of SPL, is one important aspect for the occurrence of NPF events that can enhance CCN observed in the spring and winter at SPL (Hallar et al., 2016). Other mountaintop studies that report NPF events enhancing CCN are near an anthropogenic emission source. For example, the Mt. Chacaltaya Observatory, where previous studies report 61% of events grow to CCN sizes, is located 15 km away from the city of La Paz, Bolivia (Rose et al., 2017). Mt. Tai, a mountaintop observatory in Shandong, China on the transport path of the Asian continental outflow, reports a decreased frequency of NPF events that grow to CCN sizes because of decreases in SO_2 concentrations over time, demonstrating the importance that H_2SO_4 precursors have on growing aerosols from NPF to CCN sizes (Zhu et al., 2021; Fu et al., 2008). The results from this work can be compared to other results from studies that report an enhancement of CCN due to NPF (Table 3)

16. The paper would be helped by putting the present results in perspective through a simple tabled comparison with other estimates of the contribution from NPF to CCN in the literature.

Response: This is a great idea. Table 3 in the edited manuscript now includes results from other studies that detail the enhancement of CCN due to NPF events. We limited the table to observational studies that explicitly report an enhancement factor of CCN due to NPF. A reference to the table is made on lines 396-397.

Addition: The results from this work can be compared to other results from studies that report an enhancement of CCN due to NPF (Table 3)

Additions:

Site	Authors	Environment	Time Period	NPF Frequency	Contribution of NPF to CCN
Storm Peak Laboratory, Steamboat Springs, CO, USA	Hirshorn et al., 2022	Mountaintop	2006 - 2021	50%	1.36 enhancement in winter, 1.54 enhancement in spring
Mt. Chacaltaya Observatory, Bolivia	Rose et al., 2017	Mountaintop	2012	Boundary layer: 48% Free troposphere: 39%	Boundary layer: 67% of events enhance CCN Free troposphere: 53% of events enhance CCN
Vienna, Austria	Dameto de España et al., 2017	Urban	2014 - 2015	13%	14 days display 1.43 enhancement
University of Crete at Finokalia, Crete, Greece	Kalkavouras et al., 2019	Coastal	2008 - 2015	162 episodes	1.29 – 1.77 enhancement
Polarstern Research Vessel near Svalbard, Norway	Kecorius et al., 2019	Polar	2017	4 events analyzed	Enhancement factor 2-5
Iberian Peninsula, Spain	Rejano et al., 2021	One urban site, one mountaintop site	2018-2019	Urban: N/A Mountaintop: N/A	Urban: N/A Mountaintop: 1.75

35 sites worldwide	Ren et al., 2021	Multiple Sites Urban and Remote	Varied	N/A	Urban: 3.6 enhancement Remote: 1.8 enhancement

Table 3: Details of multiple studies that find the enhancement of CCN by NPF using observational data. For a study to be included on this list, a enhancement percentage or factor of CCN due to NPF must be calculated.

17. Lines 397-398 – Scale is an important factor when comparing remote regions with areas of strong anthropogenic influence. There has been a reasonable amount of work examining NPF in the Arctic (a remote region). I think before concluding with this statement, you should look at some related literature: for example, Nieminen et al. (ACP, 2018; https://doi.org/10.5194/acp-18-14737-2018); Abbatt et al. (ACP, 2019; https://doi.org/10.5194/acp-19-2527-2019), and references therein.

Response: The authors thank the reviewer for providing these papers. We agree that the statement made should be reworded after examining the literature to place an emphasis that SPL's remote location does display differences from remote locations in polar regions. One of the big takeaways is that NPF in polar regions relies heavily on DMS from the ocean (natural influence) while our continental site might rely on power plants (anthropogenic influence). The sentence on Lines 461-463 was changed.

Addition: Similar enhancements of CCN in remote, continental regions, such as SPL, may require an anthropogenic source of NPF precursors to grow to sizes relevant to CCN formation.