

Dear Prof. Hui Su,

We would like to gratefully acknowledge the time and effort that you and the Referees have dedicated to providing valuable feedback on our manuscript, "Rapid growth and high cloud-forming potential of anthropogenic sulfate aerosol in a thermal power plant plume during COVID lockdown in India." The overall assessment of our manuscript by the Referees is positive, encouraging publication, while still providing us with valuable suggestions for further improvement.

While we have been able to meticulously incorporate the changes to reflect most of the suggestions provided by the Referees, we would like to point out that the suggestions and comments pertaining to providing additional measurement data, beyond and above the campaign we carried out, are not practically feasible, i.e., performing a similar scale campaign again at a background site and/or after lockdown is not possible. We do understand and appreciate that the additional data requested would have been immensely helpful. Even so, the scientific conclusions we have presented based on the available dataset are scientifically robust and comprehensive. Based on the suggestions of the Referees, we modified the manuscript to reflect this study as a particular case study, for which follow-up studies could provide further detailed insights into the results from the remaining campaign period.

The point-by-point and detailed Responses (in *Italic text* as AR) to the Referees' Concerns (RC) are given below. The important, convincing, and compelling summary of our revised work for your quick read-through in strong support of our claims is as follows:

- *Our observations highlight a unique new particle formation and rapid growth event under the conditions of reduced anthropogenic emissions during the lockdown over the tropical coastal environment of Chennai, India. We report an event of NPF and rapid particle growth that strongly coincided with high particulate sulfate mass fraction, which appears to be produced and grown by the condensation of H₂SO₄ resulting from the oxidation of the SO₂ plume emitted by a large coal-fired power plant located 200 km south of the observational site.*
- *The sulfate-rich particles exhibited strong cloud-forming potential over a varying range of supersaturations, with unusually high CCN fractions and κ values higher than those recorded at other times in this location.*
- *The hygroscopicity of these aerosols is more influenced by chemical composition than particle size, indicating enhanced CCN activity due to increased sulfate fraction. These measurements of aerosol composition, growth rate, CCN number concentration, and hygroscopicity are useful as datasets to evaluate the impact of altering aerosol properties on cloud and precipitation-forming processes under less polluted environment.*
- *We emphasize the importance of increased aerosol mass burden through new particle formation for framing policies for PM_{2.5} reduction plans in polluted coastal clusters.*

Thus, based on the above points and after implementing the comments from the Referees, we are confident that our manuscript certainly merits reconsideration for resubmission/publication in *Geophysical Research Letters (GRL)*.

Sachin S. Gunthe

On behalf of all the co-authors.

Reviewer #1:

This article shows a case study of the rapid particle growth strongly coincided with high sulphate fraction, mainly discuss the emitted by the large coal-fired power plant in this region, key parameters such as NPF growth rate, CCN activation, and hygroscopicity evolution has been researched, but the further investigation seems lack of evidence, the author should provide more proof to convince the reader with a clear discussion, it should be prepare a minor revision before acceptance.

We thank Referee #1 for the positive assessment of our manuscript and for providing valuable suggestions, which have greatly helped us to improve the quality of our manuscript. Below we answer the minor comments suggested by Referee #1.

[RC1.1]

L110-111: The spring festival in China is also a chance to shut down the human activity to research the lockdown period. Refers to:

a) Wang, Y., Li, Z., Wang, Q. et al., Enhancement of secondary aerosol formation by reduced anthropogenic emissions during Spring Festival 2019 and enlightenment for regional PM_{2.5} control in Beijing, Atmospheric Chemistry and Physics, 21, 915-926, 2021.

b) Christensen, M. W., Gettelman, A., Cermak, J., Dagan, G., Diamond, M., Douglas, A., Feingold, G., Glassmeier, F., Goren, T., Grosvenor, D. P., Gryspeerdt, E., Kahn, R., Li, Z., Ma, P. L., Malavelle, F., McCoy, I. L., McCoy, D. T., McFarquhar, G., Mülmenstädt, J., Pal, S., Possner, A., Povey, A., Quaas, J., Rosenfeld, D., Schmidt, A., Schrödner, R., Sorooshian, A., Stier, P., Toll, V., Watson-Parris, D., Wood, R., Yang, M., and Yuan, T.: Opportunistic experiments to constrain aerosol effective radiative forcing, Atmos. Chem. Phys., 22, 641-674, 10.5194/acp-22-641-2022, 2022.

[AR1.1]

We would like to thank Referee #1 for the suggestion. In the revised manuscript, the change is made, and references are cited appropriately.

[RC1.2]

L130: "sulfate-rich particles" more evidence should be provide, such as ACSM or AMS results. "high hygroscopic growth" is there any measurement of hygroscopic such as H-TDMA or other instruments, detail information should add.

[AR1.2]

It may kindly be noted that the chemical composition measurements presented in this study are performed using ACSM. Furthermore, the high sulfate fraction observed during the growth event is evident from the high sulfate fraction (~53%), indicating sulfate-rich particles. An H-TDMA instrument was not deployed in the campaign. The hygroscopicity parameter, kappa (κ), presented in the manuscript, is derived based on size-resolved CCN measurements. We modified the sentence as follows in the revised manuscript:

"The rapidly growing sulfate-rich particles were observed to have high hygroscopicity and the potential to serve as CCN for cloud formation."

[RC1.3]

Do you mean the baseline of background concentration of this area, in this meaning, you should measure on the remote site which represents the background intensive mixing atmosphere.

[AR1.3]

During the measurement campaign, the concentrations of the various pollutants were much lower in this region owing to the COVID-19-induced lockdown. Various studies, as also listed in the paper, have found that during the lockdown the concentration of various pollutants reached that of the background level. We clarified this point in the revised manuscript as below:

“The observations and analysis presented herein provide a unique opportunity to examine the sensitivity of CCN to new particle formation and growth due to SO₂ emissions from a coal-fired power plant under relatively cleaner conditions for tropical India, thereby establishing a baseline for compare-and-contrast to the usually prevailing conditions of heavy pollution.”

[RC1.4]

hygroscopicity parameter (κ) retrieved by CCN should be take carefully, refer to:

F. Zhang, Y. N. Li, Z. Li, R. J. Li, L. Sun, C. Zhao, P. C. Wang, Y. L. Sun, X. G. Liu, J. X. Li, P. R. Li, G. Ren, and T. Y. Fan., Aerosol hygroscopicity and cloud condensation nuclei activity during the AC3Exp campaign: implications for cloud condensation nuclei parameterization. Atmos. Chem. Phys., 14, 13423–13437, 2014.

[AR1.4]

This has been corrected with proper referencing.

[RC1.5]

One simulation was performed for each ACSM measurement instance for the given meteorological data. How to explain this? Just one trajectory been calculated or each?

[AR1.5]

One simulation was performed for each ACSM data point. So, we have as many simulations as ACSM measurement instances. This point is clarified in the revised supplementary section, and we thank Referee #1 for pointing this out.

[RC1.6]

The DU used to show the column total effective sickness unit, how to illustrate the ground and divide the upper air is a question, if you have the ground base measurement is a good verification.

[AR1.6]

The satellite imagery is used in the manuscript only to qualitatively show the influence of the SO₂ plume emitted from the Neyveli power plant on the measurement site and nearby area. For this reason, we believe that the total SO₂ column density is the best available choice, particularly in the absence of ground-based measurement, which we understand is a drawback.

[RC1.7]

Why to limit the model top in 100 m? more transport from upper layer could cause significant impact on the concentration, should also take into consideration.

[AR1.7]

We performed simulations using different model top heights now (up to 1500 meters; to depict a typical boundary layer) and found similar results. We also noticed that previous literature mentions that 100-meter height above ground level is the best representation of the back trajectory analysis for the qualitative estimation of wind pattern/direction. The plot for 1000 m AGL is added in the supplementary figure of the manuscript, as suggested by the Referee.

[RC1.8]

Diurnal variation should be drawn by every daily average plot, not just time series to see the evolution of the PM/BC concentration in different days.

[AR1.8]

As pointed out by the Referee, the diurnal variations are calculated using every daily average plot and not as a time series. The diurnal variation is shown in the inset of Fig. S1.

[RC1.9]

Assuming the density of 1.1 kg m^{-3} , Give reference measured nearby in this area or other research support this assumption.

[AR1.9]

This typo is corrected. We used the density of 1.1 g cm^{-3} , and an appropriate reference has been cited.

[RC1.10]

The nucleation mode, accumulation mode and coarse mode may be more precise to lead the understanding. How about the N_{25-50} nm variation?

[AR1.10]

To report the measurements for the nucleation mode, it is always appropriate to have measurements below 10 nm, which, unfortunately, we do not have in this field campaign. Thus, to be consistent with the standard definition and bring out the distinctive behaviour of the aerosol size distribution, we used the following diameter ranges, N_{10-25} , N_{50-100} , $N_{100-1000}$, which also helps account for different-sized particles in the Aitken and accumulation modes. The variation in N_{25-50} is similar to particles in N_{10-25} and N_{50-100} size ranges.

[RC1.11]

The pollutant transport in the area is also important, not only discuss the local emissions.

[AR1.11]

We modified the sentence as shown below. Thank you for the suggestion.

“The observed weak average diurnal variation (Fig. S1) in BC concentrations during the lockdown, however, appeared to be due to the regional background rather than persisting local

BC emissions. Wildfires and open burning are major primary sources of atmospheric BC, and their widespread occurrences result in transport and persistent presence, even in remote marine and continental boundary layer, where low local emissions are anticipated.”

[RC1.12]

It seems the wind change from north to south is the key factor influence the particle number concentration in the Fig.2g.

[AR1.12]

As the Referee rightly points out, this aspect is now emphasized in the manuscript, and the modified sentence is shown below:

“From midnight till noon, the chemical composition of NR-PM₁ was dominated by organic aerosols (Org) followed by sulfate (SO₄), ammonium (NH₄), and nitrate (NO₃) (Fig. 2a, c; Org: 50%, SO₄: 35%; NH₄: 10%, and NO₃: 5%), but following the abrupt transition of air mass from north-westerly to southerly direction, the sulfate concentration increased 1.5-2-fold (Fig. 2a, c).”

[RC1.13]

Fig.4: (d) "The different marker shapes represent the time before (circle), during (square), and after (triangle) the particle growth event". This plot only separates the observation into daylight time and nighttime, this could also be related to the boundary layer evolution, contributed to the particle growth is not appropriate here.

[AR1.13]

The plot shows the variation in CCN number concentration throughout the event, and it does not intend to separate the observation into daylight and nighttime. The event was observed during the day, and we believe that the boundary layer evolution may not have played a pronounced role in new particle formation.

[RC]

Atmospheric aerosols play an important role in climate change by scattering the radiation directly or acting as cloud condensation nuclei. New particle formation (NPF) is one of the major sources of aerosol particles. COVID lockdown provided an ideal opportunity to understand the NPF and its role in CCN activity in scenario of the restricted anthropogenic emissions. This manuscript investigated the NPF event with rapid growth during the COVID lockdown. This rapid growth event was found to be associated with the SO₂ plume from power plant. The manuscript was well written. I believe that this work can be considered for publication after the following comments have been addressed.

[AR]

We would like to thank Referee #2 for the positive evaluation of our manuscript, encouraging publication, and providing insightful comments, which have significantly helped improve our manuscript.

Major comments:

[RC2.1]

This study focused on only one NPF case during the lockdown period. The authors elucidated that it is a unique NPF and rapid growth event. Does it mean that this NPF can only occur during the lockdown period with reduced anthropogenic aerosol emission? Are there other NPF events during the measurements at the station (during the lockdown or non-lockdown period)? I suggest to give some more discussions. Some comparisons, e.g. unique NPF events v.s. other NPF events during lockdown, NPF during lockdown v.s. NPF during non-lockdown, are needed to demonstrate that this NPF event during lockdown period is unique. The comparisons of NPF related parameters, e.g. formation rate, growth rates, are needed. Moreover, are there days when the site is controlled by the air masses from power plant, but it is non-NPF event day? What are the characters of NPF events during the non-lockdown period (business-as-usual scenario)?

[AR2.1]

We thank Referee #2 for these insights and for raising the critical point about the NPF event observed at this site during the lockdown. The measurements are campaign-based, and this site has not been characterized by comprehensive data sets in the past. What we have generally observed/hypothesized, based on other similar locations and our limited/quasi-continuous data is that NPF and subsequent growth events are triggered and sustained by organic precursors during the business-as-usual scenario. We also observed a few NPF events post-lockdown but with a low sulfate fraction, indicating that organic precursors trigger those events. That is why we are presenting this study as a unique case study where the role of SO₂ emission from a power plant plume is the primary reason for this NPF and subsequent growth. During the business-as-usual NPF events outside of the lockdown, we do not expect to have such a high hygroscopicity parameter, as observed in this case study. We report this unique event, highlighting the role of particle composition on CCN activity under relatively cleaner conditions.

[RC2.2]

I suggest to calculate the hygroscopicity kappa based on the chemical composition data and compare it with kappa from CCN-SMPS system. It will give the evidences that how the particle number size distribution and chemical composition affect the CCN activity.

[AR2.2]

We thank Referee #2 for the suggestion. We carried out this analysis and found a qualitative agreement with the kappa values. The aerosol size range in this study for the size-resolved CCN measurements and ACSM measurements greatly differs, and additionally the conditioning of the aerosol during the two measurements is very different. Therefore, the additional analysis along these lines did not yield significant scientific information, and the decision was thus made to omit it from the manuscript.

[RC2.3]

Section 3 Results and Discussions: It is hard for readers to follow the discussions in this section. I suggest to divide this section into sub-section (e.g. section 3.1, section 3.2, section 3.3) and give the titles for each sub-section.

[AR2.3]

In response to the suggestion of the Referee, we prepared a version of the manuscript in this format and circulated it to co-authors. The feedback was that the sub-sections were too short and interrupted the narrative of the short manuscript in GRL format. If the Editor and Referee continue to encourage subsections, however, we would be happy to prepare the manuscript in this format.

Minor comments:

[RC2.4]

Line 1-4: The title is too long. I suggest to simplify the title.

[AR2.4]

We modified the manuscript title to make it more precise and concise.

[RC2.5]

Line 51-52: To prove this conclusion, the comparisons, e.g. "NPF in the SO₂ plume during lockdown period v.s. NPF in the SO₂ plume during non-lockdown period" and "NPF in the SO₂ plume v.s. NPF in the non-SO₂ plume", are needed.

[AR2.5]

As mentioned above (please see response [AR2.1]), we intend to report a unique NPF and growth event in this manuscript, highlighting the role of particle composition on CCN activity under relatively cleaner conditions. The intention is not a comparison between lockdown vs. non-lockdown conditions. The original wording in the manuscript was confusing. For clarity, the manuscript text is modified to clearly report it as a case study. The sentence is rephrased as follows:

“Here, based on measurements of cloud condensation nuclei (CCN) activity and chemical composition of atmospheric aerosols during the lockdown, we report an episodic event showing rapid growth and high hygroscopicity of new aerosol particles formed in the SO₂ plume from a large coal-fired power plant. These sulfate-rich particles had high CCN activity and number concentration, indicating high cloud-forming potential.”

[RC2.6]

Line 58: Plain language summary is too long for the wider readers. I suggest to shorten the plain language summary.

[AR2.6]

We thank the Referee for this suggestion. In terms of journal guidance to authors, the current summary is 143 words, and journal's rules allow a plain language summary of up to 200 words.

[RC2.7]

Line 105-106: "as evident in decreasing..."?

[AR2.7]

We apologize for the grammatical error and have now corrected it.

[RC2.8]

Line 143-144: I suggest to start the section 2.1 with the description of the location of site

[AR2.8]

We thank the Referee for the suggestion. Section 2.1 of the manuscript, titled 'Sampling Site,' outlines the measurement site description as shown below:

"The measurements of various aerosol properties were carried out with a dedicated inlet protected from insects and other blockages. The instruments were housed in a temperature-controlled laboratory at the Indian Institute of Technology (IIT) Madras (12.99 °N, 80.23 °E; 14 m above sea level), Chennai. The city of Chennai is the fifth most populous city in India with more than 10 million inhabitants. The measurement site experiences a tropical hot and humid climate, with moderate temperatures during the measurement period (11 April 2020 to 6 May 2020)."

[RC2.9]

Line 155: "ITT Madras": the full name is needed when you use the abbreviation at the first time.

[AR2.9]

The full name of the institute has been introduced in the preceding lines in the manuscript and reproduced below:

"The instruments were housed in a temperature-controlled laboratory at the Indian Institute of Technology (IIT) Madras (12.99 °N, 80.23 °E; 14 m above sea level), Chennai."

[RC2.10]

Line 157: 15 minutes is not a high time resolution.

[AR2.10]

We understand the reviewer's concern and removed "high" from the sentence.

[RC2.11]

Line 180: "BC" should be "BC".

Line 199: "the UV-Vis"?

Line 376: "business-as-usual"

Line 276: "airmass" should be "air mass".

[AR2.11]

We corrected the errors pointed out by the Referee.

[RC2.12]

Line 209: section 2.8: I suggest to merge the section 2.6 and 2.8 as both the STILT and HYSPLIT model are the Lagrangian Transport models.

[AR2.12]

We thank the Referee for the suggestion. We consulted with co-authors, and in the end, we suggest to keep the two models separate in presentation. The reasoning is that the methodology used for both models is distinct and, specifically, we used different meteorology in the STILT compared to the HYSPLIT analysis. In addition, these two models distinctly demonstrate the scientific finding which helped us to provide better scientific interpretations.

[RC2.13]

Line 225: Why do you fix the top of model layer at 100 m? I think the top of model layer should be 10000 m and you can output the layer of 100 m a.g.l.

[AR2.13]

Please see response [AR1.7].

[RC2.14]

Line 248: Fig. 2g does not correspond to the conclusion of this sentence.

[AR2.14]

We apologize for the inconsistency here and thank the Referee for pointing it out. The correct figure corresponding to this sentence is Fig. 2b.

“New particle formation (NPF) on this day was triggered after sunrise in the continental air mass (Fig. 2b) with a sustained and rapid increase in total particle number concentrations (N_{Tot}) and geometric median diameter (D_{gmd}) until 11:30 hrs.”

[RC2.15]

Line 297-299: Even though few studies reported such high growth rates, the references about high growth rates need to be cited and discussed further here.

[AR2.15]

Appropriate references have been cited, and the difference in triggering chemistry (organics usually, sulfate in this case) has been discussed, as shown below:

“Very few studies have reported such a significant and rapid growth in D_{gmd} during a growth event (Guo et al., 2014; Pierce et al., 2012). A study in Beijing reported that a high amount of freshly nucleated nanoparticles from organic precursors during a clean period often precedes episodes of high pollution due to the growth of these nanoparticles to bigger sizes leading to increased aerosol mass burden. It was also observed that the local sulfate and nitrate emissions further enhanced the rapid growth of these newly formed particles (Guo et al., 2014).”

[RC2.16]

Line 315: H₂SO₄ is also an important precursor of nucleation, such as H₂SO₄-NH₃ nucleation, H₂SO₄-DMA nucleation. In fact, H₂SO₄ may play a minor role in particle growth especially in the large size range.

[AR2.16]

We agree with the Referee that H₂SO₄ is an important nucleation precursor responsible for new particle formation, as observed in our study. We hypothesize that sustained growth of the newly formed particles is due to H₂SO₄ condensation and heterogeneous uptake of SO₂ under humid conditions. Lower concentrations of pre-existing particles further enhanced the chances of H₂SO₄ condensing onto the newly formed particles, contributing to particle growth. Previous studies have also reported similar findings under clean marine conditions as well as polluted continental locations (References provided in the manuscript). This explanation is included in the revised manuscript as follows:

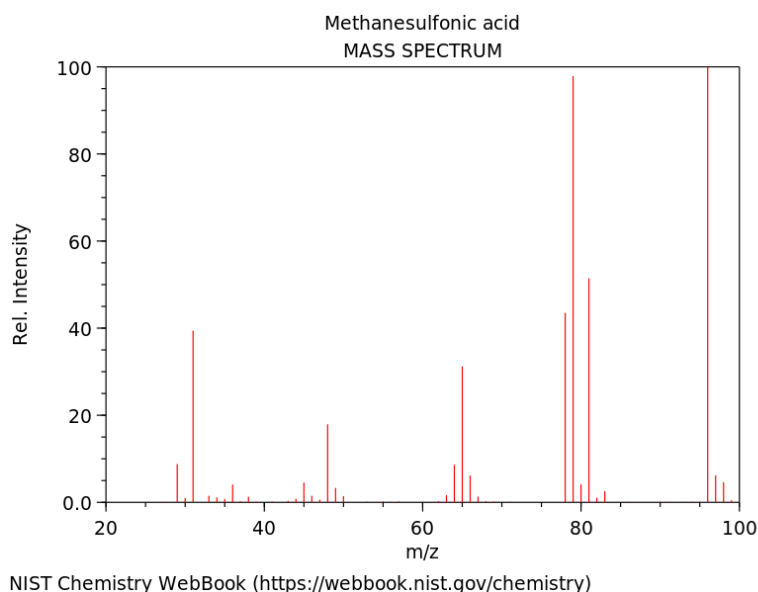
“However, it is critical to note that condensation of H₂SO₄ alone may not be sufficient to explain the exceptionally high growth rates of the newly formed particles (~20 nm/h for particles above 50 nm in diameter) observed during this event. The size-growth of the “sulfate-rich” particles may have also resulted from increasing heterogeneous uptake of SO₂ with increasing RH and particle diameter as the day progressed, especially after 12:00 hrs (Fig. 2b, f). Several previous studies at urban sites have indicated that sulfate increased quicker with elevated RH and particle diameter than organics (G. Wang et al., 2016; Zheng et al., 2016).”

[RC2.17]

Line 325-326: Can ACSM measure the MSA? Why does the m/z 79 represent the MSA?

[AR2.17]

Yes, ACSM can measure MSA signals. The standard spectral signal of MSA includes very high signals at m/z 79 and 96, as the compound fragments in mass spectrometry analysis (figure attached). Since an m/z 96 peak can also indicate other sulfate compounds, a high m/z 79 is taken as a proxy for MSA instead.



[RC2.18]

Line 370-371: Again, some evidences (or comparisons) are needed to demonstrate this NPF event is unique.

[AR2.18]

As we have mentioned, we are reporting a unique NPF and growth event in this manuscript, highlighting the role of particle composition on CCN activity under relatively cleaner conditions. The manuscript is clarified as follows:

“The sulfate-rich particles exhibited strong cloud-forming potential over a varying range of supersaturations, with unusually high CCN fractions and κ values higher than those recorded at other times for this location. The hygroscopicity of these aerosols is found to be more influenced by the chemical composition than particle size, indicating enhanced CCN activity due to increased sulfate fraction.”

“Our results, to the best of our knowledge, are the first direct observational evidence signifying the role of SO_2 emitted from a coal-fired power plant in strongly enhancing the CCN activity of aerosol particles.”

[RC2.19]

Line 376-377: Evidences are needed to prove this conclusion. I wonder if the pre-existing aerosol can really suppress the NPF in power plant plume during non-lockdown period. Previous studies also found the strong NPF in power plant plume.

[AR2.19]

Pre-existing particles can suppress new particle formation by scavenging the condensable vapors available for nucleation. A large number of pre-existing particles provides a larger surface area for condensation of H_2SO_4 vapors; thus, nucleation is limited. Appropriate references have been provided to support this conclusion in the manuscript. The manuscript is clarified as follows:

L348-355: “The reduced number concentration of pre-existing aerosol particles due to reduced anthropogenic emissions during lockdown seems to have enhanced the new particle formation and growth observed during this study. A lesser concentration of pre-existing aerosol particles implies a lower condensation sink (CS) for H₂SO₄ condensation, leading to enhanced ultrafine particle formation through nucleation. It has been previously reported that only a small fraction of the H₂SO₄ condenses onto the pre-existing particles, while 90% condenses onto the newly-formed particles under cleaner marine conditions (Stevens et al., 2012). This leads to the sustained growth of nucleated particles to bigger sizes, as observed in the present study.”

[RC2.20]

Line 435: The description on MODIS data is needed.

[AR2.20]

As per Referee’s suggestion, the text in the caption of figure 1 is revised as follows:

“ The daily Level 2 (L2) data for Aerosol Optical Depth (AOD) with a spatial resolution of 10 km x 10 km was retrieved from the atmospheric aerosol product (MOD04_L2) of the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor on board NASA's Terra satellite, which records the aerosol optical thickness over the land surfaces and oceans, globally.”

[RC2.21]

Line 443: Figure 1d only show the BC data in one wavelength.

[AR2.21]

The Aethalometer measures light attenuation due to particle deposition at 7 different wavelengths; however, the standard for calculating BC mass concentration is calculated from the attenuation observed/ measured at 880 nm, which we have reported here. Appropriate references citing this method are cited in the manuscript. The manuscript is clarified as follows:

" The instrument measures aerosol spectral light absorption, which has been converted to BC mass concentrations using a mass absorption cross-section at 880 nm (Drinovec et al., 2015).

[RC2.22]

Line 461: What does the total condensation sink mean? Is it the condensation sink of all the gas vapors? The methods of calculating the condensation and coagulation sink need to be described in section 2 or SI.

[AR2.22]

The total condensation sink is a measure of the lifetime of sulfuric acid and other similar volatility vapors against condensation on pre-existing aerosol particles. The condensation sink (CS) determines how rapidly vapor molecules will condense onto existing particles and depends strongly on the shape of the particle size distribution. The method for calculation of condensation and coagulation sinks are now included in the supplementary information.

[RC2.23]

Figure 1: I suggest to show the time series of SO₂ as well.

[AR2.23]

Although we agree with the Referee that a time series of SO₂ data would have been beneficial as additional ancillary data for supporting our presented scientific conclusions, gaseous SO₂ measurements were not part of the campaign's data set.

[RC2.24]

Figure 4b, c: what do the dots in Fig. 4b c represent?

Font size is too small in some figures (e.g. Fig. 3d, e, f, Fig. 4). Please unify the font size.

[AR2.24]

The dots in Fig. 4b and c represent the kappa values calculated from size-resolved CCN measurements at different times during the NPF and growth event for supersaturation levels of 0.15% and 0.8%, respectively. We revised the description in the caption of Fig. 4 for clarity. Based on the Referee's suggestion, we unified the font size in the figure panels.

[RC]

This study conducted a field measurement to explore the potential of newly formed particles to act as CCN during the COVID lockdown in a big city Chennai, India. They proposed that the reduced aerosol concentrations during COVID lockdown have caused high new particle growth rates, and the elevated sulphate fractions in the newly formed particles have exhibited high CCN potential. They further proposed that the elevated sulphate fraction is from the power plant plume, indicating the importance of anthropogenic emission during the clean periods. However, the current dataset cannot support the main conclusions. In terms of the measurements, no measurement for H₂SO₄ or even direct SO₂ concentration is available; the new particle composition is measured by ACSM which theoretically cannot measure particles smaller than 40-50 nm; BC data is not directly related to NPF or CCN. In terms of the effect of COVID lockdown, the key characteristics of NPF, particle composition, and CCN are only acquired during the lockdown period (actually only one NPF event is observed during the one month) but no comparison before or after the lockdown periods. In terms of the influence from the power plants, Fig 3e indicates that the high SO₂ is not caused by the power plant emission. Thus, I do not recommend publication unless more convincing data is provided and the following questions are well addressed.

[AR]

We would like to thank Reviewer #3 for the detailed and insightful reading of our manuscript and the subsequent points raised here are well-taken. The following points have helped us immensely to improve our manuscript further.

- *For H₂SO₄ and/or SO₂, please see [AR2.23].*
- *For ACSM, we apologize for the confusion here; we did not intend to mention that new particle composition is measured by ACSM. We meant that the particle composition above 40-50 nm was dominated by sulfate during the new particle growth event. The text is modified as follows:*

“However, the chemical composition of the newly formed particles in the Aitken mode cannot be ascertained as the ACSM is unsuitable for characterizing particles smaller than ~50 nm. The increase in sulfate concentration recorded by the ACSM, which coincided with the increase in particles in N₅₀₋₁₀₀ nm and N₁₀₀₋₁₀₀₀ nm size ranges, mainly represents the increasing sulfate mass fraction in particles between ~70-700 nm, as the collection efficiency of the instrument is optimum in this range (P. S. K. Liu et al., 2007). As evident from Fig. 2b, for a brief period at the start of NPF event, we observed a bimodal aerosol number size distribution with modal diameters of <30 nm and ~100 nm.”

- *Our BC data is measured throughout the campaign, and we have used the BC data to demonstrate the minimum local anthropogenic emissions during the lockdown as compared to the business-as-usual scenario. Supplementary Fig. 1 shows the BC concentrations before and during the lockdown. Also, we provided comparisons of mass fractions of various chemical species during the lockdown and business-as-usual time in supplementary Fig. 3. In this regard, the text is clarified as follows to call the reader's attention:*

“Black carbon (BC) concentrations, an indicator of combustion and pollution (Putaud et al., 2004), persistently remained less than 5 µg m⁻³ during the measurement period due to the COVID-induced lockdown (average concentration 1.5±0.7 µg m⁻³; Fig 1d).

In contrast, the average concentrations were ~3-4 times higher ($5.4 \pm 1.9 \mu\text{g m}^{-3}$; Fig. S1) and exhibited a pronounced diurnal variation for a similar duration before lockdown during the business-as-usual scenario, suggesting elevated local emissions. The observed weak average diurnal variation (Fig. S1) in BC concentrations during the lockdown, however, appeared to be due to the regional background rather than persisting local BC emissions. Wildfires and open burning are major primary sources of atmospheric BC, and their widespread occurrences result in transport and persistent presence, even in remote marine and continental boundary layer (Schill et al., 2020a), where low local emissions are anticipated. Looking more in detail at the period of interest, the weak correlation between SMPS-PM_{0.5} and BC ($R^2=0.18$, Fig. S2) indicates reduced combustion-related anthropogenic activities, such as traffic and industrial emissions.”

“To explore the possible origin of the observed sulfate particles, we calculated the associations of the BC+Org fraction (proxy of local combustion emissions) and SO₄ fraction with NR-PM₁+BC (i.e., PM₁) concentrations. We found a negative association of BC+Org with PM₁, whereas SO₄ was positively associated with PM₁, which is usually opposite for the business-as-usual scenario (Fig. S3).”

Major comments:

[RC3.1]

The conclusion for COVID lockdown period should be accompanied by comparisons with pre-lockdown or after-lockdown periods. (1) Line 51: "contrary to normal conditions of heavy pollution", the article did not provide any useful observational data about the situation during heavy pollution; (2) Line 67-69: the author should provide the measured or estimated H₂SO₄ concentration before and during COVID lockdown to support this conclusion. (3) Line 69-71: the author should provide particle composition and CCN activity before and during COVID lockdown to support this conclusion. (4) Line 316-317: "NPF and growth are significantly enhanced" compared to what? What was the situation before the lockdown? And there is only one NPF event during the lockdown period, how could say "significantly enhanced?" (5) Figure 2c, provide the composition of new particles before the lockdown periods.

[AR3.1]

Line 51: - We provided the BC and ACSM data regarding aerosol loading before and during the lockdown in supplementary figures and also discussed the same in the manuscript. Supplementary Fig. 1 clearly shows the BC concentrations before and during the lockdown. Further, we provided comparisons of mass fractions of various chemical species during the lockdown and business-as-usual scenario in Supplementary Fig. 3. In this regard, the text is clarified as follows to call the reader's attention:

“Black carbon (BC) concentrations, an indicator of combustion and pollution (Putaud et al., 2004), persistently remained less than $5 \mu\text{g m}^{-3}$ during the measurement period due to the COVID-induced lockdown (average concentration $1.5 \pm 0.7 \mu\text{g m}^{-3}$; Fig 1d). In contrast, the average concentrations were ~3-4 times higher ($5.4 \pm 1.9 \mu\text{g m}^{-3}$; Fig. S1) and exhibited a pronounced diurnal variation for a similar duration before lockdown during the business-as-usual scenario, suggesting elevated local emissions. The observed weak average diurnal variation (Fig. S1) in BC concentrations during the lockdown, however, appeared to be due to

the regional background rather than persisting local BC emissions. Wildfires and open burning are major primary sources of atmospheric BC, and their widespread occurrences result in transport and persistent presence, even in remote marine and continental boundary layer (Schill et al., 2020a), where low local emissions are anticipated. Looking more in detail at the period of interest, the weak correlation between SMPS-PM_{0.5} and BC ($R^2=0.18$, Fig. S2) indicates reduced combustion-related anthropogenic activities, such as traffic and industrial emissions.”

Line 67-69:- Please see [AR2.23].

Line 69-71 and Fig2c:- Again, having all the data presented here before lockdown would have been immensely helpful; however, such data sets do not exist. Therefore, we modified the text to avoid any confusion and present this study as a case study, presenting a unique observation NPF and subsequent growth in powerplant plume as evident from high sulfate fraction, which is very unusual during an NPF event (generally marked by a higher organic fraction). See [AR2.5] for text revisions.

Line 316-317:- We revised the manuscript for more clarity, as follows:

“The reduced number concentration of pre-existing aerosol particles due to reduced anthropogenic emissions during lockdown seems to have enhanced the new particle formation and growth observed during this study. A lesser concentration of pre-existing aerosol particles implies a lower condensation sink (CS) for H₂SO₄ condensation, leading to enhanced ultrafine particle formation through nucleation. It has been previously reported that only a small fraction of the H₂SO₄ condenses onto the pre-existing particles, while 90% condenses onto the newly-formed particles under cleaner marine conditions (Stevens et al., 2012). This leads to the sustained growth of nucleated particles to bigger sizes, as observed in the present study.”

As the lockdown was phased out, we observed a few more NPF and growth events, which were not marked with high sulfate fraction and hence were not affected by power plant plume emissions. Since those events are scientifically different, we omitted them from the focus of the present study.

[RC3.2]

The authors did not mention that ACSM is not suitable to measure particles smaller than 40-50 nm. For Figure 2a, at least before 12:00, the composition cannot represent new particle composition. The 50% transmission efficiency of particles in ACSM aerodynamic lens is ~40-70 nm. For particles smaller than this diameter, the sampling efficiency is very low, combining with the small mass of the nanoparticles, ACSM cannot be used for sub-40 nm particle measurements. For the size distribution presented in Figure 2b, we could see that before 12:00, there are two modes of the particles, and the larger mode should dominate the ACSM-measured mass instead of the smaller NPF mode (by the way, we usually use log scales instead of linear scale for the color bar, then you can see the larger mode is very high). To quantitatively check how much of the measured mass is from the NPF mode, the authors can multiply the measured size distribution with ACSM aerodynamic transmission efficiency (Liu et al., 2007), and then convert the number distribution to volume distribution.

[AR3.2]

We do understand Referee's point about the ACSM particle size limitation. We now explicitly mention in the manuscript (supplementary information as well as main text) that the collection efficiency of ACSM below 40 nm is very low and that it optimally measures particles in the size range of 70-700 nm, along with a proper reference. Please see below the revised text. We are in agreement with the Referee that the second mode (larger particle mode) cannot be attributed to sub-40 nm particles for the ACSM-derived chemical composition measurements. That is the reason we see the dominance of the organics (as generally observed for larger particles), potentially because the sulfate-rich particles were in the sub-40 nm mode and were not captured by the ACSM. This is further corroborated by the fact that the sulfate fraction in the ACSM starts increasing at around 12:00 hrs when the particle number size distribution mode reaches ~70 nm. This point also coincides with the sharp reduction in the ORG-SUL ratio. We also added these points in the manuscript (see below). We prepared the volume size distribution in response to the suggestion by the Referee, but after review with the co-authors this plot did not provide additional scientific value. Finally, as suggested, we changed the scale in Fig. 2b to a log scale.

“However, the chemical composition of the newly formed particles in the Aitken mode cannot be ascertained as the ACSM is unsuitable for characterizing particles smaller than ~50 nm. The increase in sulfate concentration recorded by the ACSM, which coincided with the increase in particles in N_{50-100} nm and $N_{100-1000}$ nm size ranges, mainly represents the increasing sulfate mass fraction in particles between ~70-700 nm, as the collection efficiency of the instrument is optimum in this range (P. S. K. Liu et al., 2007).”

[RC3.3]

Line 300-319: The "sulfate-rich" particles are very likely caused by the increasing heterogeneous uptake of SO₂ with increasing RH and dp rather than H₂SO₄ condensation.

[AR3.3]

Based on Referee's suggestion, we included this point in the revised manuscript. Previous studies have reported the growth of particles due to SO₂ uptake under high humidity conditions; however, heterogeneous uptake of SO₂ can be only partially responsible for the growth of particles in our case, mainly due to the absence of freshly emitted sulfate from local sources. Also, the increase in sulfate concentration occurred after the abrupt transition in wind direction, which clearly indicates that the change in wind direction likely caused the high sulfate concentration and subsequent particle growth. The text is revised as follows:

“The size-growth of the "sulfate-rich" particles may have also resulted from increasing heterogeneous uptake of SO₂ with increasing RH and particle diameter as the day progressed, especially after 12:00 hrs (Fig. 2b, f). Several previous studies at urban sites have indicated that sulfate increased quicker with elevated RH and particle diameter than organics (G. Wang et al., 2016; Zheng et al., 2016).”

[RC3.4]

Firstly, from Figure 2f, RH is always higher than 50% and increases even higher after 12:00, and particle diameter increase to above 50 nm after 12:00. These all benefits the heterogeneous uptake of SO₂. Many previous studies in polluted urban sites have indicated that with elevated RH and particle diameter, sulfate increased quicker than that of organics (Wang et al., 2016; Zheng et al., 2016). For composition before 12:00, new particles are

smaller than 50 nm so the ACSM measurements are not representative. (2) Secondly, the growth rates of the new particles are so quick (~ 20 nm/h) for above-50 nm particles, this makes us wonder how high H₂SO₄ concentration can support these high growth rates for such large particles. That must be extremely high! Especially considering the solar radiation after 12:00 may decrease. The authors should at least use an H₂SO₄ proxy to estimate its condensational growth rate.

[AR3.4]

Please see [AR3.3]. In addition, as mentioned in the studies suggested by the Referee, studies in "polluted" environments attribute the heterogeneous uptake of SO₂ to local sources, where there is no sufficient time for oxidation. Whereas, in our case, the source is regional; hence we believe that heterogeneous SO₂ uptake may be only partially responsible for rapid particle growth. This is further supported by the fact that the modal diameter of the measured size distribution does not seem to increase with increasing RH.

[RC3.5]

The data in Figure 3 cannot support that the new particle formation is significantly influenced by the power plant plume. (1) According to Figure 3e, SO₂ column density is high for the whole map, this could not be the influence from the power plant. The reason for the risen of SO₂ should be further explored. (2) According to Figure 3f, the plume concentration has decreased at least ~ 2 orders of magnitude after the 200 km's long-range transport and the plume concentration is only $1\text{e-}5$ mg/m³ at the observation site, which is too small to exert big difference considering the background SO₂ column is so high as shown in Figure 3e.

[AR3.5]

We thank the Referee for this observation regarding Figure 3. This comment pointed out the need to improve the figure further. The concentration appeared to be high throughout the region because of the small color scale we had used for SO₂ column density (0-0.5 DU). However, we have now modified Fig. 3e by increasing the scale range to 0-1 DU, which provides better contrast and clearly indicates the influence of the SO₂ plume from the power plant on the observational site. Also, it must be noted that satellite data for a brief period of time (a day in our case) may be based on a single or only a few orbital measurements and hence be noisy.

We agree with the Referee that the plume concentration appears to have decreased by approximately two orders of magnitude by the time it reached the measurement site; however, compared to clean background conditions, the plume concentrations remain high. It is important to note that the conversion of SO₂ to sulfate through oxidation will result in net decrease in SO₂ concentration. Also, the role of meteorology in this particular event looks pivotal [see AR3.3 and AR3.4]. For clarification, the text is revised as follows:

"On the growth event day, particularly during 08:00-18:00 hrs, the STILT sensitivity analysis, TROPOMI satellite imagery, and HYSPLIT dispersion modeling results showed strong sensitivity of the measurement site to SO₂ emissions originating from the south/southeast direction (Fig. 3b, e, and f). These observations support the scenario that the SO₂ plume emitted from the Neyveli power plant situated southeast of the observational site (Fig. 3f) contributed to the high sulfate in the particulate matter.

[RC3.6]

Figure 4 is hard to read and is somehow misleading. (1) Figure 4a, the representative diameter is very misleading, ACSM can not measure 10 nm particle composition. The representative diameter should be the measured volume median diameter of ACSM after multiplying with real size distribution and transmission efficiency in ACSM, as calculated in question 2. (2) For Figure 4b-c, it seems κ decreases with the increase of particle diameter, which is opposite with the authors' statement. What is the black line represents? (3) Figure 4e, increasing heterogeneous uptake with dp and RH can explain this.

[AR3.6]

We thank the reviewer for pointing this out, and we have now rectified this. In particular, the color scale representing the sulfate fraction has been removed from Fig. 4a-c because we agree that ACSM measurements cannot be taken as representative of the chemical composition of particles smaller than ~40 nm. We use the representative diameter from the SMPS measurements. The intention is to show the evolution of aerosol number size distribution throughout the NPF and growth event in Fig. 4a. Fig. 4b-c are the same as Fig. 4a, but for CCN number size distribution obtained from size-resolved CCN measurements for specific supersaturations of 0.15% and 0.8%, respectively. For clarity in response to the Referee comment, the caption is revised as follows:

“Various characteristic aerosol properties derived using size-resolved CCN, ACSM, and SMPS measurements on the day of the NPF and rapid particle growth event at Chennai. (a) Particle number size distribution (normalized particle concentration in cm^{-3} plotted against diameter in nm). The individual aerosol number size spectra obtained from SMPS measurements plotted represent the time of the day before, during, and after the particle growth event. (b) Same as (a) but for CCN number size distribution derived for 0.15% effective supersaturation. (c) Same as (a) but for CCN number size distribution derived for 0.79% effective supersaturation. The markers in (b) and (c) represent the hygroscopicity parameter κ , calculated from size-resolved CCN measurements for each scan of 0.15% and 0.79% effective supersaturation, respectively, during the entire NPF and growth event.”

Here, we want to emphasize the influence of increasing sulfate mass fraction on the CCN activity of particles. As evident from Fig.2b, the shape of the CCN-number size distribution and the critical dry diameter at a low supersaturation did not vary much with increasing sulfate mass loading; however, κ increased from 0.15 to ~0.55, indicating a strong role of chemical composition in causing higher hygroscopic growth. Further, at the higher supersaturation of 0.8%, we see an increased number of CCN-active particles in the Aitken mode as the sulfate fraction increases, reinforcing the significant role of particle chemistry at higher hygroscopicity in this study.

[RC3.7]

Line 233-247 and Line 333-346, the discussions on BC are too much. It is just an assistant indicator of the local combustion sources and does not directly support any main conclusions. It is directly related to neither NPF nor CCN.

[AR3.7]

For clarification, the discussion on BC is to iterate and emphasize that local emissions were low during the time of the NPF and growth event reported here. Based on the Referee's suggestion, we reduced the BC discussion in the revised manuscript.