Rapid growth and high cloud forming potential of anthropogenic sulfate aerosol during the Covid lockdown in India: Changes in the production and properties of cloud con-densation nuclei (CCN) during heavily polluted compared to relatively cleaner condi-tions

Aishwarya Singh[1], Subha S Raj[2], Upasana Panda[3], Snehitha Kommula[1], Christi Jose[1], Tianjia Liu[4], Shan Huang[5], Basudev Swain[6], Mira L. Pöhlker[2], Ernesto Reyes Villegas[7], Narendra Ojha[8], Aditya Vaishya[9], Alessandro Bigi[10], Ravikrishna R[1], Qiao Zhu[11], Liuhua Shi[12], James Allen[7], Scot T. Martin[4], Gordon McFiggans[7], Meinrat O. Andreae[2], Ulrich Poschl[2], Hugh Coe[7], Federico Bianchi[13], Hang Su[2], Vijay P. Kanawade[14], Pengfei Liu[15], and Sachin S Gunthe[16]

[1]Indian Institute of Technology Madras
[2]Max Planck Institute for Chemistry
[3]CSIR Institute of Minerals and Materials Technology
[4]Harvard University
[5]Institute for Environmental and Climate Research, Jinan University, Guangzhou 511443, Guangdong, China
[6]Institute of Environmental Physics, Department of Physics, University of Bremen
[7]University of Manchester
[8]Physical Research Laboratory (PRL)
[9]National University of Ireland, Galway
[10]University of Modena and Reggio Emilia
[11]Emory University
[12]Department of Environmental Health, Harvard T H Chan School of Public Health, Boston, MA, USA
[13]University of Helsinki
[14]University of Hyderabad
[15]School of Earth and Atmospheric Sciences, Georgia Institute of Technology
[16]Environmental and Water Resources Engineering Division, Department of Civil Engineering, Indian Institute of Technology Madras, Chennai 600036, India.

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Abstract

Covid lockdown presented an important opportunity to study relatively cleaner conditions in India. The complex factors of power production, industry, and transportation could be more carefully dissected because of the extreme reduction in the influence of the latter two emission sources. Measurements of cloud condensation nuclei (CCN) activity and other chemical properties of atmospheric aerosols showed that newly formed aerosol particles were produced in the SO_2 plume from a large coal-fired power plant, contrary to normal conditions of heavy pollution. The sulfate-rich particles had high CCN activity and

number concentration, indicating high cloud-forming potential. Examining the sensitivity of CCN properties under relatively clean conditions over India provides important new constraints on the perturbations of past and future climate forcing by anthropogenic emissions. Because most sensitive regime of aerosol climate forcing on cloud development is the midpoint of relatively clean conditions afforded by the Covid lockdown between background and polluted conditions.

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[1]EE Division, Dept. of Civil Engineering, Indian Institute of Technology Madras, Chennai, India

[2]Laboratory for Atmospheric and Climate Sciences, Indian Institute of Technology Madras, Chennai, India

[3]Department of Environment and Sustainability, CSIR – Institute of Minerals and Materials Technology, Bhubaneswar, India

[4]Department of Earth and Planetary Sciences, Harvard University, Cambridge, MA, U.S.A.

[5]Institute for Environmental and Climate Research, Jinan University, Guangzhou, Guangdong 511443, China

[6]Institute of Environmental Physics, Department of Physics, University of Bremen, NW1, Otto-Hahn-Allee 1, 28359 Bremen, Germany

[7]Multiphase Chemistry Department, Max Planck Institute for Chemistry, Mainz, Germany

[8]Department of Earth and Environmental Sciences, School of Natural Sciences, University of Manchester, Manchester, U.K.

[9]Space and Atmospheric Sciences Division, Physical Research Laboratory, Ahmedabad, India

[10]School of Arts and Sciences and Global Centre for Environment and Energy, Ahmedabad University, Ahmedabad, India

[11]Dipartimento di Ingegneria 'Enzo Ferrari', University of Modena and Reggio Emilia via Vivarelli, Modena, Italy

[12]Dept. of Chemical Engineering, Indian Institute of Technology Madras, Chennai, India.

[13]Gangarosa Department of Environmental Health, Rollins School of Public Health, Emory University, Atlanta, Georgia, U.S.A.

[14]National Centre for Atmospheric Science, University of Manchester, Manchester, U.K.

[15]John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA, U.S.A.

[16]Scripps Institution of Oceanography, University of California San Diego, La Jolla, CA, U.S.A.

[17]Department of Geology and Geophysics, King Saud University, Riyadh, Saudi Arabia

[18]Institute for Atmospheric and Earth System Research/Physics, University of Helsinki, Helsinki, Finland

[19]Center for Earth, Ocean, and Atmospheric Sciences, University of Hyderabad, Hyderabad, India

[20]School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA, U.S.A.

*Correspondence to: Sachin S. Gunthe (s.gunthe@iitm.ac.in) and Pengfei Liu (pengfei.liu@eas.gatech.edu)

Abstract

Covid lockdown presented an important opportunity to study relatively cleaner conditions in India. The complex factors of power production, industry, and transportation could be more carefully dissected because of the extreme reduction in the influence of the latter two emission sources. Measurements of cloud condensation nuclei (CCN) activity and other chemical properties of atmospheric aerosols showed that newly formed aerosol particles were produced in the SO_2 plume from a large coal-fired power plant, contrary to normal conditions of heavy pollution. The sulfate-rich particles had high CCN activity and number concentration, indicating high cloud-forming potential. Examining the sensitivity of CCN properties under relatively clean conditions over India provides important new constraints on the perturbations of past and future climate forcing by anthropogenic emissions because most sensitive regime of aerosol climate forcing on cloud development is the midpoint of relatively clean conditions afforded by the Covid lockdown between background and polluted conditions.

Plain language summary

Atmospheric aerosol particles play an essential role in cloud formation and precipitation. During the Covid lockdown in India, the ambient concentrations of atmospheric pollutants, including aerosols, dropped significantly. To understand the human impact on cloud formation under relatively cleaner conditions, we performed specialized measurements of cloud forming potential and chemical properties of aerosols in Chennai. The Neyveli coal-fired power plant (3390 MW) located 200 km south of the observational site was fully operational during lockdown. When the power plant plume intersected the observation site, there were

high sulfate concentrations and associated new particle formation, which was followed by rapid particle growth. Unlike during the polluted conditions, the reduced local aerosol concentration provided a smaller surface area for sulfuric acid condensation, leading to the growth of newly formed particles. Because of the elevated sulfate concentration, atmospheric aerosol particles exhibited high cloud forming potential, otherwise lowered under polluted conditions. Our results show that cloud-forming particle concentrations can be highly sensitive to anthropogenic emissions in relatively cleaner environments representing important implications for understanding human influences on climate. Paradoxically, heavy pollution under normal economic activity can reduce the cloud-forming activity of individual particles by diluting the sulfate mass from power plant emissions.

1. Introduction

The accurate quantification of aerosols responsible for climate-forcing relative to the pre-industrial era is crucial for estimating the climate impacts caused by current anthropogenic enhancement M. Andreae & Rosenfeld, 2008Pachauri & Reisinger, 2007(;). The aerosol climate forcing associated with changes in cloud albedo depends not only on the enhancement of anthropogenic aerosol concentration but also on the baseline concentration, which is largely determined by natural emissions K. S. Carslaw et al., 2013Liu et al., 2021(;). Compared with heavily polluted conditions, the cloud condensation nuclei (CCN) concentration, the cloud droplet concentration, and the cloud albedo effect is more sensitive to the increase of anthropogenic emissions under relatively cleaner conditions. Therefore, the uncertainty in climate forcing estimates is often linked to a lack of knowledge about aerosol properties under relatively clean environments, as well as background conditions before industrialization M. O. Andreae, 2007(), and our knowledge in this regard largely relies on chemistry-climate modeling rather than measurements, because of the pervasive pollution conditions that typically prevail today, especially in India. Given the highly non-linear response of cloud properties to aerosol concentration, additional measurements in relatively cleaner conditions can help unravel how precursor emissions, aerosol formation and growth, and aerosol properties including number concentration and chemical composition impact cloud condensation nuclei concentrations and climate forcing through aerosol-cloud interactions Kenneth S. Carslaw et al., 2017K. S. Carslaw et al., 2013Fan et al., 2016Hamilton et al., 2018Liu et al., 2021Rosenfeld et al., 2014(; ; ; ; ;). During the Covid lockdown in India, an interesting scientific opportunity presented itself during the transition from heavily polluted conditions to relatively cleaner conditions.

The pandemic outbreak of coronavirus (COVID-19) affected more than 10 million people across India Gunthe & Patra, 2020Gunthe et al., 2020Krishnamoorthy et al., 2020(; ;). To contain the domestic spread of COVID-19, India imposed a nationwide lockdown beginning on 25 March 2020, limiting the economic activity of 1.3 billion people and leading to large reductions in anthropogenic emissions until 31 May 2020 Chatterjee et al., 2021(). The lockdown

caused a drastic reduction in major local anthropogenic emissions, as evident in decreasing concentrations of major pollutants, including PM_1, $PM_{2.5}$, and BC (Fig. 1a,b) across India Goel et al., 2020C. D. Jain et al., 2021S. Jain & Sharma, 2020Karuppasamy et al., 2020Kumar et al., 2020Kumari & Toshniwal, 2022Lokhandwala & Gautam, 2020Navinya et al., 2020R. P. Singh & Chauhan, 2020V. Singh et al., 2020Vadrevu et al., 2020(; ; ; ; ; ; ; ; ; ;). While there was an overall change from heavily polluted conditions to relatively cleaner conditions, the power industry continued to operate normally during this lockdown period. This setup offered the interesting scientific opportunity to isolate the effects of this industry from the normally entwined activities of other industries and transportation, as well as to study the sensitivity of CCN properties in the important transition regime of relatively cleaner conditions for the India region, which has not been possible under the conditions of heavy pollution that have prevailed before and after the Covid lockdown. The impact of SO_2 emissions from a coal-fired power plant on aerosol processes under relatively cleaner conditions could be studied and compared to the business-as-usual scenario.

In this context, the coastal city of Chennai experienced a substantial reduction in the local anthropogenic pollution S. Jain & Sharma, 2020(). On the other hand, the Neyveli coal-fired power plant located 200 km south of the observational site continued full operation during the Covid lockdown. This power plant does not seem to have effective emissions controls for SO_2 and other pollutants. The Neyveli Thermal Power Station, located in the southern state of Tamil Nadu in India, has an installed capacity of 3390 MW as of April 2021 and produces 2740 MW of electricity from its three operational units, emitting 299 kt SO_2 a^{-1} Fioletov et al., 2020().

Here, we report comprehensive aerosol measurements from Chennai during the Covid lockdown. When the power plant plume intersected the observation site, there were high sulfate concentrations and associated new particle formation, which was followed by rapid particle growth. These sulfate-rich particles had high hygroscopic growth and the potential to serve as CCN for cloud formation. The observations and analysis presented herein provide a unique opportunity to examine the sensitivity of CCN to new particle formation and growth due to SO_2 emissions from coal-fired power plant under relatively cleaner conditions for tropical India, thereby establishing a baseline for compare-and-contrast to the normally prevailing conditions of heavy pollution. The analysis includes a comparison to those conditions to understand better the particular role of industrial power production in that more complex scenario of heavy pollution from other industries as well as transportation.

2. Materials and Methods

The sampling site and measurements are introduced in this section. A detailed description of the measurement site, instruments, calibration procedure, brief results, meteorological measurements, and data analysis are described in supporting information (SI).

2.1 Sampling site

The measurements of various aerosol properties were carried out with a dedicated inlet, which was protected from insects and other blockages. The instruments were housed in a temperature-controlled laboratory at the Indian Institute of Technology Madras (12.99^{o}N, 80.23^{o}E; 14 meters 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). The average aerosol and meteorological parameters (arithmetic mean±standard deviation) obtained during the measurements are summarized in Table S1.

2.2 PM_1 Chemical composition measurements

The chemical composition measurements of non-refractory particulate matter with diameters smaller than 1 µm (NR-PM_1) were carried out using an Aerosol Chemical Speciation Monitor (ACSM; Aerodyne Research Inc., U.S.A.) Ng et al., 2011(), which was housed in the dedicated laboratory at IIT Madras. The measured chemical composition included organics (Org), sulphate (SO_4), nitrate (NO_3), ammonium (NH_4), and chloride (Cl) at a high time resolution of 15 minutes and unit mass sensitivity. Maintenance and calibration of the instrument was performed before the beginning of the campaign and once during the campaign as per the detailed protocol Freney et al., 2019().

2.3 $PM_{0.5}$ mass derivation

The aerosol number size distribution was measured using a Scanning Mobility Particle Sizer (SMPS, T.S.I. Inc), which comprised of a 3082 Electrostatic Classifier with 3081 Long Differential Mobility Analyzer (L-DMA), and a 3772 butanol-Condensation Particle Counter (CPC). The measurements were carried out with an aerosol flow rate of 1.0 lpm, covering the size range of 9.82 – 414.2 nm. The mass concentration of total $PM_{0.5}$ was derived from SMPS aerosols number size distribution, assuming the particle density of 1.1 kg m^{-3} DeCarlo et al., 2004().

2.4 Size resolved CCN measurements

The size-resolved CCN measurements were carried out using a continuous flow stream-wise thermal gradient CCN counter (CCNC, Model 100, Droplet Measurement Technologies, U.S.A.) operated in synchronization with an SMPS (T.S.I., 3081 DMA+3776 CPC), in addition to the one mentioned above. The CCNC supersaturation (S) was systematically cycled through 7 different values between 0.06 and 0.8%, which are determined by the temperature gradient (T) in the CCNC column. The critical supersaturation (S_c) and activation diameter (D_{50}) pair, derived from size-resolved CCN measurements, was used to calculate the hygroscopicity parameter kappa ( ). The calibration of the CCN counter was carried out as described in Rose et al., 2008() and the estimation of the hygroscopicity parameter ( ) is based on Petters & Kreidenweis, 2007().

2.5 Black carbon measurements

Black carbon (B.C.) mass concentrations were measured at 1-minute time resolution using the dual-spot model AE33 Aethalometer (Magee Scientific, Berkeley, U.S.A.) at seven different wavelengths ( ) with a flow rate of about 2 lpm. 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(). Measurement by the dual-spot Aethalometer (AE33) is known to minimize the artifacts associated with multiple scattering and filter loadings.

2.6 STILT model simulations

We performed simulations using the STILT (Stochastic Time-Inverted Lagrangian Transport) model (version 2.0 Fasoli et al., 2018()) to compute the transport of an ensemble of approximately 2000 air particles to the receptor site for 120 hours backward in time, using Global Data Assimilation System (GDAS) data with a spatial resolution of $0.5°\times0.5°$. One simulation was performed for each ACSM measurement instance for the given meteorological data. We used the STILT footprints to estimate the average sensitivity of the receptor site to nearby potential sources for high sulphate condition.

2.7 TROPOMI satellite data

The TROPOspheric Monitoring Instrument (TROPOMI), boarded on the Sentinel-5 Precursor satellite, provides high resolution spectral observations of SO_2 by using ultraviolet (UV)–visible (Vis) part of the electromagnetic spectrum. The instrument gives daily global coverage data in the UV–Vis region with a horizontal spatial resolution of 3.5 km $\times$ 5.5 km. The algorithm for the operational TROPOMI SO_2 retrieval is described in Theys et al., 2017(). The level-2 data used in this study can be accessed through the website (http://www.tropomi.eu/data-products/sulphur-dioxide). As per the recommendations mentioned in the product Readme File for SO_2 data, only pixels having qa-value > 0.5 should be used. Therefore, only the pixels for which the data quality is high (qa-value > 0.5), are considered for this study. Instead of using mol/m^2, the unit of the tropospheric SO_2 column number density is converted to Dobson Units (DU). The data is then regridded into a lat/lon grid to combine the data from multiple orbits from one day into a single daily grid by using properely weighted area average to calculate the value for each grid cell.

2.8 HYSPLIT dispersion model analysis

We used the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) by NOAA's Air Resources Laboratory, for simulating the atmospheric dispersion of the plume arising from the Neyveli thermal power plant on the day of the rapid particle growth event. The HYSPLIT model is described in detail elsewhere Rolph et al., 2017Stein et al., 2015(;). NCEP/NCAR Global Reanalysis Data for different pressure levels, having 2.5 degree latitude-

longitude global grids, was used as input meteorology for dispersion modelling. The required meteorology files were obtained from the ARL server, which contains 3-d, gridded, ARL/HYSPLIT-format compatible files. The HYSPLIT dispersion model was run using the Neyveli power plant (11.553°N, 79.441°E) as the source, emitting SO_2 continuously at the rate of 822 kg/day from stack height of 30-50 m AGL. The emission rate and stack description for the power plant are taken from Fioletov et al., 2020(). A simple forward dispersion simulation from the continually-emitting source was carried out for 24 hours starting at 30 April 2020 12:30 UTC (18:00 IST), and ending at 01 May 2020 12:30 UTC (18:00 IST; roughly coinciding with the end of particle growth event). For simplicity, a plain terrain was considered, with no wet or dry deposition of the particles emitted from the stack. The vertical top of the model layer was fixed at 100 m AGL and the 24-hour average concentrations were obtained within this layer as the model output.

3. Results and Discussions

The time series and other characteristic properties of NR-PM_1, SMPS-$PM_{0.5}$, BC, and $PM_{2.5}$ measured at Chennai during the COVID-19 lockdown period are shown in Fig. 1. These measurements were carried out between 11 April 2020 and 6 May 2020. Black carbon (BC) concentrations, an indicator of combustion and pollution Putaud et al., 2004(), persistently remained less than 5 µg m^{-3} during the lockdown (average concentration 1.5±0.7 µg m^{-3}; Fig 1d). The observed weak average diurnal variation (Fig. S1) in BC concentrations appears to be due to the regional background rather than persistent local BC emissions. In contrast, the BC concentrations for a similar duration before lockdown during the business-as-usual scenario exhibited pronounced diurnal variation, and average concentrations were ~3-4 times higher (5.4±11.9 µg m^{-3}; Fig. S1) than during the lockdown period, suggesting elevated local emissions. 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 the reduced combustion-related anthropogenic activities during the lockdown. Interestingly, on 1 May, a day dominated by prevalent southerly winds (Fig. 1c) and relatively low average BC concentration (1.9±0.7 µg m^{-3}; Fig. 1c,d), NR-PM_1, and SMPS-$PM_{0.5}$ mass concentrations reached ~40 µg m^{-3} (Fig. 1e), with corresponding $PM_{2.5}$ concentration reaching ~40-50 µg m^{-3} as evident in Fig. 1d (marked by the golden shaded region). The strong correlation between SMPS-$PM_{0.5}$ and NR-PM_1 (Fig. 1f; R^2=0.8; N=92) and a relatively weak correlation between SMPS-$PM_{0.5}$ and BC (R^2=0.45; Fig. S2) indicates the relatively low contribution of local combustion-related anthropogenic sources to the total aerosol mass burden on 1 May.

Rapid particle growth on this day was triggered after sunrise in the continental air mass (Fig. 2g) with a sustained and rapid increase in total particle number concentrations (N_{Tot}) and geometric median diameter (D_{gmd}) until 11:30 hrs. At around 11:30 hrs, however, a break in the growth event was noticed, potentially caused by an abrupt transition of air masses (Fig. 2g), and as a result,

the aerosol size distribution appears as a broken banana-shaped aerosol growth as reported by previous studies Nilsson & Kulmala, 1998Wehner et al., 2007Yu et al., 2014(; ;). The rapid aerosol particle growth event at this site indicates the greater availability of low-volatility vapors for aerosol nucleation and growth under cleaner conditions, which pre-existing aerosol particles would otherwise scavenge. This is evident in Fig. 2e, showing a low condensation sink of 5×10^{-3} s^{-1} before the rapid growth event. Remarkably, a decrease in the Org:SO_4 ratio from ~4 before the onset of the growth event to as low as ~0.1 by the end of the particle growth event indicated a significant contribution of sulfate to the aerosol growth (Fig. 2f). This is in contrast with previous observations showing the dominance of organic fraction in nanoparticle growth Mohr et al., 2019Paasonen et al., 2010Riipinen et al., 2012Smith et al., 2008Stolzenburg et al., 2018Yli-Juuti et al., 2020(; ; ; ; ;).

An abrupt transition of the air mass from continental (northwest wind) to the south/southeast wind occurred at around 11:30 to 12:00 hrs (Fig. 3), indicating the strong influence of the SO_2 plume emitted from coal fired power plant (discussed below). The plumes from coal-fired power plants generally exhibit higher concentrations of SO_2 than in a typical boundary layer Stevens et al., 2012(). As a result of the transition period, there was a transient decrease in the number of particles and mass concentrations; however, the rapid growth of the newly formed particles was observed to continue after 12:00 hrs until 18:00 hrs, exhibiting the typical characteristics of particle growth (Fig. 2b). From midnight till noon, the chemical composition of NR-PM_1 was dominated by organic aerosols (Org) followed by sulfate (SO_4), ammonium (NH_4), and nitrate (NO_3) (Fig. 2a, c; Org: 50%, SO_4: 35%; NH_4: 10%, and NO_3: 5%), but following the abrupt transition of air mass from northwesterly to southerly direction, the sulfate concentration increased 1.5-2 fold (Fig. 2a, c). This implies that the sulfate-enriched air mass resulted in an exceptionally high fraction of sulfate in the total particle mass (Fig. 2c) during the particle growth. The airmass back-trajectory analysis, STILT modeling, satellite imagery (Fig. 3), and in-situ wind direction measurements (Fig. 1c) distinctly show the influx of the SO_2 plume emitted from the Neyvely coal-fired power plant during the aerosol growth event, suggesting the transport of sulfate-rich particles potentially formed during the plume transport. In the downwind SO_2 plumes from coal-fired power plants, aerosol can undergo a rapid transformation and aging Stevens et al., 2012().

The number concentration for particles in the size range of 10-25 nm diameter ($N_{10\text{-}25\ nm}$) started increasing approximately at 08:00 hrs local time, reaching as high as 10,000 cm^{-3} within about 20 minutes, contributing 77% of N_{Tot}, which likewise increased by five times (from ~2500 cm^{-3} to ~13000 cm^{-3}; Fig. 2d). The increase in $N_{50\text{-}100}$ nm and $N_{100\text{-}1000}$ nm was observed later during the aerosol growth event, consistent with previous studies reporting NPF and particle growth events Kulmala et al., 2004Pierce et al., 2012Westervelt et al., 2014(; ;). The coagulation sink (CoagS) is about two orders of magnitude lower than condensation sink (CS), indicating a less significant loss of newly formed particles to pre-existing aerosols, which was also low under lockdown-

induced cleaner conditions. The CS sharply increased by about a factor of 4 after 12:00 hrs due to a sharp change in air mass and as particles grew to CCN-active sizes (i.e., 50 nm). The growth event, which continued until 18:00 hrs, weakened in terms of the particle concentration with N_{Tot}, $N_{\text{10-25 nm}}$, $N_{\text{50-100 nm}}$, and $N_{\text{100-1000 nm}}$ reaching 5000 cm^{-3}, <50 cm^{-3}, 2500 cm^{-3}, and 4000 cm^{-3}, respectively. The Org:SO_4 ratio started to increase at 18:00 hrs, coinciding with the end of the growth event,. A consistent increase in GMD was noticed during the event, increasing from ~20 nm to as high as ~160 nm, not as a constant growth but as the appearance of a new mode around noon. Very few studies have reported such a significant and rapid growth in GMD during a growth event Guo et al., 2014Pierce et al., 2012(;).

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 SO_4 fraction with NR-PM_1+BC (i.e., PM_1) concentrations. We found a negative association of BC+Org with PM_1, whereas SO_4 is positively associated with PM_1, which was the opposite during the business-as-usual scenario (Fig. S3). These results, combined with the persistent high SO_4 concentration after the wind direction change, indicate that the observed SO_4 was not emitted from local sources, and the observations may represent broader regional conditions. The STILT model simulation, satellite images, and back trajectory analyses further suggest that during the condensational growth of the aerosol particles, the observational site received prevalent winds dominated by the SO_2 plume from the Neyveli coal-fired power plant (Fig. 3a). On the day of the growth event, particularly during 08:00-18:00 hrs, the STILT model results and dispersion modeling (Fig. 3a, f) showed strong sensitivity to SO_4 emissions originating from the south/southeast of the observational site (Fig. 3e). This supports the scenario that the SO_2 plume emitted from the Neyveli power plant located approximately 200 km southeast of the observational site (Fig. 3f) contributed to high sulfate in particulate matter. The SO_2 emitted from the power plant is oxidized to H_2SO_4, which can condense onto newly formed aerosol particles. New particle formation and growth are significantly enhanced during the reduced polluted conditions during this study because of the lockdown, as pre-existing aerosol particles during polluted conditions may provide a large surface area for H_2SO_4 condensation and subsequent particle growth Stevens et al., 2012().

Another possible mechanism of growth event and high particulate sulfate at a marine location could be triggered by the precursors resulting from biogenically emitted dimethyl sulfide (CH_3SCH_3; DMS.) oxidation, and methane sulfonic acid (CH_3SO_3H; MSA.) has been used as a proxy using online mass spectrometry measurements Huang et al., 2017Huang et al., 2018(;). The biogenically emitted DMS from open oceans undergoes oxidation to H_2SO_4 and MSA, leading to aerosol nucleation and growth. Given the limited unit mass resolution data of the ACSM, we did not find any evidence of enhancement in m/z79, representing MSA. Since the MSA:nssSO_4 ratio can be quite low (<0.1) even in clean marine conditions Schill et al., 2020a() the lack of detectable MSA is not conclusive evidence against a contribution from biogenic DMS. However, to produce such

a high sulfate concentration would require an even greater concentration of DMS, which would exceed the highest DMS concentration ever reported, and therefore such a possible source originating from oceanic surface processes is ruled out in this case.

It is also noted that BC concentrations recorded during the aerosol growth event were not especially low, representing non-zero continental emissions. Wildfires and open burning are major and primary sources of atmospheric BC, and their widespread occurrences result in transport and persistence presence of BC, even in remote marine and continental boundary layer Schill et al., 2020b(), where low local emissions are anticipated. The lack of a diurnal variation in BC concentrations indicates a low level of local, particularly traffic sources. Regional anthropogenic activities could be a significant source of BC even during the lockdown, but do not seem to contribute significantly during our measurements, owing to the lack of local emissions. This clearly indicates the reduced emissions of local anthropogenic origin; however, it substantiates the possibility of SO_2 emissions from the power plant, which emits 376 tons SO_2 per year. In contrast, the efficient combustion in power plants releases relatively little BC and organics (photographs of the Neyveli power plant show no black smoke emanating from the stacks). This would result in sulfate/BC ratios much higher than from diesel trucks, biomass burning, etc.

Nevertheless, regardless of the potential sources accountable for the rapid growth of particles, we observed strong implications for the cloud formation, as evident from the evolution of the aerosol number size distribution and CCN concentration (Fig. 4). At the onset of the growth event, particles were very small (D_{gmd} 15 nm) with <10% sulfate fraction and accumulation mode particles already present in the atmosphere dominating the CCN number concentration. Thereafter, the aerosol particles exhibited rapid growth with an increase in the sulfate fraction, with D_{gmd} reaching as high as ~160 nm and a sulfate fraction >65% (Fig. 4a). At the lowest effective supersaturation (S_{eff}=0.15% and average D_{50}=138.1±14.3 nm), the shape of the CCN distribution continued to be unaffected, while the sulfate fraction increased from ~20 to 70% as the day progressed. Interestingly, the corresponding values did not exhibit a strong size dependence, but increased from the lowest value of ~0.16 to ~0.55 from morning to evening, indicating the exclusive role of changing aerosol chemical composition due to increasing sulfate fraction (Fig. 4b). At the highest S_{eff}=0.8% from the onset of the rapid particle growth, the size distribution was dominated by the accumulation mode but with high CCN number concentrations increasing in the Aitken mode (Fig. 4c). Similar observations were previously reported at an urban site in Beijing Wiedensohler et al., 2009(). The rapid aerosol particle growth, which was associated with high sulfate fraction, clearly indicated the high cloud forming ability in terms of the higher hygroscopicity parameter, particularly for Aitken and accumulation mode particles (Fig. 4d). The role of increasing diameter for higher cloud forming ability in this case strongly appeared to be related to the increase in sulfate fraction (Fig. 4e). Overall, the increasing sulfate fraction clearly shows

a positive association with incresing values.

4. Summary

In summary, our observations highlighted a unique new particle formation and rapid growth event under the reduced anthropogenic aerosol emissions during lockdown over this tropical coastal environment. We find that the rapid particle growth strongly coincided with high sulfate fraction, which in the presence of reduced anthropogenic activities appears to be either formed or grown by the condensation of H_2SO_4 resulting from the oxidation of the SO_2 plume emitted by the large coal-fired power plant located ~200 km south of the observational site. Under the business as usual scenario, such a new particle formation and growth would be suppressed due to the large pre-existing aerosol surface area from local sources for H_2SO_4 condensation and particle coagulation. The sulfate-rich particles exhibited strong cloud-forming potential over a varying range of supersaturations, with unusually high CCN fraction and values higher than the business-as-usual scenario. These measurements of aerosol composition, growth rate, CCN number concentration, and hygroscopicity may be useful datasets to evaluate the impact of altering aerosol properties on cloud and precipitation forming processes under a less polluted environment. Our results, to the best of our knowledge, are the first direct observational evidence signifying the role of coal-fired power plant emitted SO_2 in strongly enhancing the CCN activity of aerosol particles. We further emphasize the importance of increased aerosol mass burden through new particle formation to be carefully assessed while framing policies to abet the $PM_{2.5}$ reduction plans for polluted coastal clusters across India.

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Author contributions: S.S.G. conceived the idea and designed the research. A. Singh conceptualized and planned the filed campaign under the supervision

of S.S.G., and R.R.K. A. Singh led the field measurement campaign to collect the extensive filed measurement data during the COVID-19 induced lockdown with remote support from S.S.R., S.M.K., U. Panda, S.S., and C. J. V.P.K. carried out the NPF-related data analysis. V.P.K. and S.S.G. conducted NPF data interpretation with critical inputs from F.B., and A.B. U. Panda, S.M.K., S.S.G., and A. Singh conducted the ACSM data analysis. S.H., P.L., S.S.G., J.A., E.R.V., G.M., and L.S. conducted the ACSM data interpretation. A. Singh and SSG led the size-resolved CCN data analysis with help from S.S.R. A. Singh, and M.P. conducted the size-resolved CCN data interpretation with help from S.S.G. and P.L. A.V. performed the black carbon data analysis. A.V., S.S.G., V.P.K., and A. Singh conducted the black carbon data interpretation. T.L. performed the STILT simulation and data interpretation with help from P.L. and S.S.G. S.S.G. wrote the first draft and led the manuscript writing by mentoring A.Singh with valuable edits and contributions from V.P.K., NO, P.L., and R.R. during the manuscript writing. Critical inputs were provided by H.S. and F.B., with inputs and edits to the manuscript further provided by G.M., U. Pöschl, S.T.M., M.O.A., and H.C.

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Figure Caption:

Figure 1: Mass concentration of sub-micrometer particulate matter (PM_1) measured by an Aerosol Chemical Speciation Monitor (ACSM), mass concentration of $PM_{0.5}$ derived from a Scanning Mobility Particle Sizer (SMPS), black carbon concentration measured with an Aethalometer (AE33), and $PM_{2.5}$ mass concentration obtained from the United States of America Embassy in Chennai. The measurements are from the coastal city of Chennai carried out during the strictly implemented COVID-19 lockdown (25 March – 6 May 2020). The light golden shading indicates the rapid particle growth event day. (a) Map showing the location of Chennai, color scaled with MODIS L2 AOD averaged during 28 March and 5 May 2019, the time of the year equivalent to that of the COVID-19 lockdown in India in 2020. (b) Same as (a) but for the year 2020. The substantial reduction in MODIS AOD is observed over India during COVID-19 lockdown. (c) 24-hour back trajectories obtained for each hour between 05:30 hrs. and 19:30 hrs. (local time) clearly indicating strong marine influence in prevailing airmass despite the change in wind direction. The direction of hourly back trajectories is strongly consistent with the wind direction measured using the automatic weather station at measurement site. (d) Time series of Non-Refractory PM_1 (NR-PM_1) mass from ACSM, mass concentration derived from SMPS (SMPS-$PM_{0.5}$) number size distribution measurements in the size range of ~10 – 450 nm and converted to mass concentration assuming the density of 1.1 kg m^{-3}, and black carbon concentration as measured by Aethalometer in 7 different wavelengths for the campaign duration from 11 April 2020 to 6 May 2020. (e) Mass concentrations of NR-PM_1, SMPS-$PM_{0.5}$, and U.S. Embassy $PM_{2.5}$ for 8 days. The extremely high concentration observed on 1 May is highlighted in blue color. (f) Scatter plot between NR-PM_1 and SMPS-$PM_{0.5}$ for the

event day indicating the high mass concentration detected by both measurement techniques.

Figure 2: Time series of various aerosol characteristic properties observed and derived for 1 May 2020 on the day of rapid particle growth event. (a) Diel variations in chemical components in NR-PM_1 measured during the event day (b) Sub-micrometer particle number size distribution measured by SMPS showing rapid particle growth during the event (local time plotted against diameter and scaled by normalized particle number concentration). The yellow line indicates the geometric median diameter of the mode fitted to the number size distribution and the black line shows the black carbon concentration, which remained <4 µg m^{-3} throughout the event as opposed to the increasing total aerosol number concentration during the growth event, indicating the absence of local anthropogenic sources. (c) Average mass fractions of different chemical species in NR-PM_1 measured during the growth event: averaged before the growth event (left), during the growth event (center), and after the growth event (right). (d) Size segregated particle number concentration as measured by SMPS over entire measured diameter range (N_{Tot}), over 10 – 25 nm ($N_{10\text{-}25}$), over 50 – 100 nm ($N_{50\text{-}100}$), and over 100 – 1000 nm ($N_{100\text{-}1000}$). (e) Total condensation and coagulation sink during growth event day. (f) Diel variations in temperature measured at Chennai on 1 May, color scaled by relative humidity (%). On the same plot, the dark blue line indicates the diel variation in organic to sulphate ratio measured by ACSM. (g) Diel variation in wind speed. The wind speed is color coded by the wind direction. A drastic shift in the wind direction is observed just before noon coinciding with a reduced aerosol number concentration during the particle growth event.

Figure 3: Sensitivity of the measurement site with respect to nearby potential emission sources. The sensitivity presented here is derived using the Stochastic Time-Inverted Lagrangian Transport (STILT) model. (a) Sensitivity during the entire measurement period during the lockdown, (b) for 1 May when the particle growth event was observed. The STILT model was driven with meteorological data obtained from Global Data Assimilation System (GDAS) with a spatial resolution of 0.5°×0.5°. The corresponding back trajectory footprint was resampled at a finer resolution of 0.01°×0.01°. Note that the color bar is on a logarithmic scale, and (c) particle number size distribution for 1 May binned according to wind direction. The black line indicates the diameter on a logarithmic scale and the white line shows the total particle concentration corresponding to each wind direction bin. (d) SO_2 column density as obtained from TROPOMI satellite averaged over the campaign period, (e) same as (d) but averaged over 30 April and 1 May, and (f) NOAA HYSPLIT dispersion model run in forward mode showing the plume from Neyveli power plant reaching the observational site for 1 May.

Figure 4: Various characteristic aerosol properties derived using size-resolved CCN, ACSM, and SMPS measurements on the day of the particle growth event at Chennai. (a) Particle number size distribution (diameter in nm plotted

against normalized particle concentration in cm^{-3}). 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. The spectra are color coded by the sulfate fraction obtained by ACSM measurements indicating increased sulfate fraction with increase in geometric median diameter of the fitted spectrum as the day progresses. (b) Same as (a) but for CCN size number distribution derived for 0.15% effective supersaturation. (c) Same as (a) but for CCN number size distribution derived for 0.79% effective supersaturation. (d) The relation between critical dry diameter, D_{crit} (nm), and effective supersaturation estimated from size-resolved CCN measurements. The different marker shapes represent the time before (circle), during (square), and after (triangle) the particle growth event. These different markers are further color coded by the hygroscopicity parameter ( ) calculated using the individual pairs of D_{crit} (nm) and effective supersaturation on the day of the particle growth event. (e) Scatter plot between geometric median diameter of the mode fitted to the number size distributions measured by SMPS and the sulphate fraction measured by ACSM (N=42 and R^2=0.85). The dotted red line is a linear fit to the points.

Open Research

The data used in the manuscript has been deposited in an open research depository as raw data files, accessible at (https://doi.org/10.6084/m9.figshare.20418960). The aerosol size and chemical composition data, along with the meteorological data used in this manuscript, were obtained from experimental measurements and are summarised in Table S1 in Supporting Information S1. The $PM_{2.5}$ data is obtained from the US Embassy in Chennai and can be accessed through https://www.airnow.gov/international/us-embassies-and-consulates/#India$Chennai.The STILT model (Version 2, (Fasoli et al., 2018)) used in the manuscript is openly accessible at https://github.com/uataq/stilt (https://doi.org/10.5281/zenodo.1238047), while the meteorology data obtained from Global Data Assimilation System (GDAS) can be accessed at https://www.ncei.noaa.gov/products/weather-climate-models/global-data-assimilation. The dispersion modelling of SO_2 emission from Neyveli Power Plant was performed using the HYSPLIT dispersion model (Rolph et al., 2017; Stein et al., 2015), accessible at https://www.ready.noaa.gov/hypub-bin/dispasrc.pl. The SO_2 emission rate from the power plant was taken from Fioletov et al., (2020). The level-2 SO_2 satellite data was retrieved from TROPOMI using the algorithm described in Theys et al., (2017), and the data can be accessed through http://www.tropomi.eu/data-products/sulphur-dioxide. Further, hygroscopicity calculations from size-resolved CCN measurements were performed on MATLAB version R2020b (Mathworks Inc., 2020), licensed under IIT Madras and accessible at https://in.mathworks.com/products/matlab.html. The codes used for calculating and plotting data are available from the corresponding author upon reasonable request. Figures were prepared using Igor Pro version 8.03 (WaveMetrics Inc.), licensed by SSG and accessible at https://www.wavemetrics.com/downloads/filtered/Igor%20Pro%208.

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