New Particle Formation Events Observed during the COALA-2020 Campaign

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Abstract:

Aerosols play an important role in atmospheric processes influencing cloud formation, scattering and absorbing solar radiation, and play an important role in chemical reactions affecting the abundance of trace gases in the atmosphere. Ultimately aerosols affect the radiative balance of the earth modifying climate. A large fraction of aerosols is formed through chemical reactions following gas-to-particulate processes in the atmosphere: nucleation, condensation and growth. Biogenic Secondary Organic Aerosols (BSOA) are formed when plant produced volatile organic compounds (VOCs) react in the atmosphere through heterogeneous reactions. One of the highest BVOC emitting regions in the world is South-east Australia due to the high density of *Eucalyptus* species. The COALA-2020 (Characterizing Organics and Aerosol Loading over Australia) campaign worked towards a better understanding of biogenic VOCs in quasi-pristine conditions in the atmosphere and their role in particle formation.

The observations showed a highly reactive atmosphere with frequent new particle formation (NPF) occurring (50% days with data) often associated with pollution plumes. Analysis of NPF events suggested that SO_2 plumes likely triggered particle formation, while particle growth depended on available VOCs, hydroxyl radicals (influenced by relative humidity), and the presence of multiple SO_2 intrusions promoted growth of smaller clusters. Nighttime NPF events coincided with monoterpene ozonolysis. One nighttime NPF event showed potential isoprene nitrate oxidation enhancing growth, but the limited night-time data hindered conclusive interpretations. These findings highlight the significant role of biogenic VOCs, in driving NPF and SOA formation in South-east Australia, even after major wildfires. The COALA-2020 campaign provided valuable insights into local atmospheric chemistry and its potential impact on regional air quality and climate. However, longer-term observations are crucial to understand seasonal variations, trends and extreme events.

Keywords: COALA-2020; aerosols, BVOCs, NPF.

1. Introduction

Aerosols can influence our health (Annesi-Maesano et al., 2013; Shi et al., 2016) but also play an important role in regulating Earth's energy balance, the hydrological cycle and even the abundance of key chemical species in the atmosphere such as hydroxyl radical (OH) and indirectly greenhouse gases (e.g., Kerminen et al., 2012). The chemical composition, size and concentrations determine the effects on health and the environment (Liu et al., 2016b; Pope and Dockery, 2006; Ren et al., 2017). Aerosols can be directly emitted (primary aerosols) or they can be product of chemical reactions in the atmosphere (secondary aerosols) (Pöschl, 2005).

Secondary aerosols are produced via gas-to-particle transition. New Particle Formation (NPF) occurs when multiple reactions in the atmosphere create stable molecular clusters. Once the clusters are formed, they can grow through coagulation and condensation potentially resulting in cloud condensation nuclei (CCN) (Dal Maso et al., 2005; Hussein et al., 2005; Kulmala et al., 2001). Multiple factors determinate NPF in the atmosphere including atmosphere composition and boundary conditions (temperature, humidity, PBL height, turbulence) (Bousiotis et al., 2021; Wu et al., 2021a; Xu et al., 2021a). Sulfuric acid (H_2SO_4) is one of the main drivers of the nucleation process in the continental boundary layer, but it does not explain all growth and nucleation rates (Sihto et al., 2006). The presence of ammonia (NH_3), amines or ions in the atmosphere can enhance H_2SO_4 nucleation rates (Kirkby et al., 2023; Zhao et al., 2011; Zheng et al., 2015). High levels of SO_2 and Volatile Organic Compounds (VOCs) in a humid atmosphere will enhance NPF (Nestorowicz et al., 2018; Song et al., 2019, p.20; Xu et al., 2021b).

VOCs are a group of carbon-based gases emitted by biological and anthropogenic sources that are characterised by their high vapour pressure (Goldstein and Galbally, 2007; Kesselmeier and Staudt, 1999; Matsui, 2006). VOCs can undergo hydroxyl radical (OH), ozone or nitrate radical (NO3) oxidation in the gas phase, producing compounds of varying volatilities, and products with low enough volatility can contribute to NPF or partition to existing particles, resulting in particle growth.

The most common biogenic VOC (BVOC) is isoprene followed by monoterpenes. BVOCs play an important role in secondary organic aerosol (SOA) formation (e.g., Mahilang et al., 2021). VOCs have been associated with particle growth (Riipinen et al., 2012) but their role and the autoxidation mechanism was not understood until recently (Bianchi et al., 2019). Autoxidation of monoterpenes supports the particle growth process by generating highly oxygenated molecules (HOMs) via the formation of peroxy radicals (Bianchi et al., 2019; Kirkby et al., 2023; Lehtipalo et al., 2018, p.201). HOMs can be characterised as ultra-low VOCs (ULVOC) or extremely low VOCs (ELVOC) depending upon the saturation concentration at 298K.

While oxidation of monoterpenes produces higher SOA formation yields than isoprene resulting to the formation of ULVOC or ELVOC molecules during OH or ozonolysis oxidation(Friedman and Farmer, 2018; Lee et al., 2023; Luo et al., 2024; Riva et al., 2019; Zhang et al., 2018), under the right conditions, isoprene can also lead to SOA formation (e.g. Fry et al., 2018). Isoprene oxidation products can oxidise to LVOC promoting particle growth in the larger sizes (above 3 nm) (Heinritzi et al., 2020). For example, the SOA yield of isoprene oxidised with the nitrate radical at night generates several times the yield observed through the OH oxidation path (e.g. Ng et al., 2008). Recent studies suggest that in biogenic-rich regions isoprene-SOA yield in low NO_x environments can be much higher than previously reported when considering further oxidation of the products (e.g. Liu et al., 2016a), promoting the formation of extremely low or low volatility organic compounds (ELVOC and LVOC respectively) important for particle grow.

Isoprene, monoterpenes, OH and nitrate radical and ozone availability in the atmosphere are key to promoting SOA formation (e.g. Song et al., 2019). As a VOC molecule oxidises it becomes more complex and a larger oxygenated VOC (OVOC),so is less likely to be oxidised again, particularly when in the presence of other VOCs with higher OH reactivity (Kiendler-Scharr et al., 2009). HOMs are key for nucleation, but HOM production in the atmosphere is limited by other reactions/byproducts. Isoprene NPF suppression is one of those cases in which isoprene oxidation products limit the formation of C_{20} dimers and reduce the yield of highly oxygenated molecules (HOMs) in favour of weaker nucleators C_{15} (Dada et al., 2023; Heinritzi et al., 2020; Liu et al., 2016a). This effect is non-linear nor constant and will change with local conditions.

Understanding BVOC emissions and their role in SOA formation is important to accurately predict aerosol properties and their impact on climate. However, BVOC are poorly characterized under Australian conditions (Paton-Walsh et al., 2022). MEGAN emissions show south-east Australia as one of the BVOC hot-spots in the region (Guenther et al., 2012) but multiple modelling studies have shown that MEGAN emissions estimation might not represent local conditions correctly in this region (Emmerson et al., 2016, 2018, 2019). Most of the Australian forested regions are dominated by high emitting *Eucalyptus* species (ABARES, 2019; Aydin et al., 2014; Padhy and Varshney, 2005) that combined with periods of high temperature and drought stress create the conditions for high emissions/concentrations of BVOCs in the atmosphere (Emmerson et al., 2020; Fini et al., 2017; Ormeño et al., 2007). The emissions ratios of isoprene to other VOCs are poorly constrained and the local chemistry is not well understood.

The COALA-2020 campaign worked towards a better understanding of biogenic VOCs in quasi-pristine conditions in the atmosphere and their role in local atmospheric chemistry in south-east Australia. COALA-2020 was a collaborative effort between local institutions including the University of Wollongong, CSIRO, ANSTO, and the University of Sydney, and international peers from Georgia Institute of Technology, The University of California, Irvine, Nagoya University and Lancaster University. This part of the study focused on identifying and characterising NPF events after the “Black Summer” 2019-2020 Australian bushfire season. Here we focus on identifying drivers and conditions in which NPF started or were enhanced in the local environment.

2. Materials and Methods

2.1 The COALA-2020 Campaign

The COALA-2020 campaign took place at Cataract Scout camp (34°14’44” S, 150°49’26” E) located 20 km north-northwest of Wollongong on the east coast of NSW, Australia. The site is surrounded by a heavily forested area mainly stocked by Eucalyptus species (see Figure 1). North of the sampling site is a four-lane arterial road connecting the M1 motorway on the east coast with south-western Sydney. Other possible anthropogenic sources impacting the site are two underground coal mine heads, located 1.5 km to the northeast and 2.5 km to the north). Further afield sources include the Sydney suburban area (around 18 km north-west), Sydney city (45 km north), Wollongong urban area and Port Kembla steelworks in the southern part of Wollongong (28 km to the southeast).

The campaign was conducted 17 January to 23 March, 2020. The first period of the campaign (17 January to 5 February) was heavily impacted by smoke pollution from the bushfires affecting the region On 5 February A substantial rain event extinguished the fires and cleared the atmosphere of residual smoke pollution (Mouat et al., 2022; Simmons et al., 2022). The smoke pollution period has been removed from the analysis presented here as we focus on understanding atmospheric processes during more normal conditions. Thus, this paper presents the analysis of BVOCs alongside anthropogenic emissions and their role in NPF during *the second part of the COALA-2020 ambient measurements campaign running from 5 February 5th until March 17th 2020.*

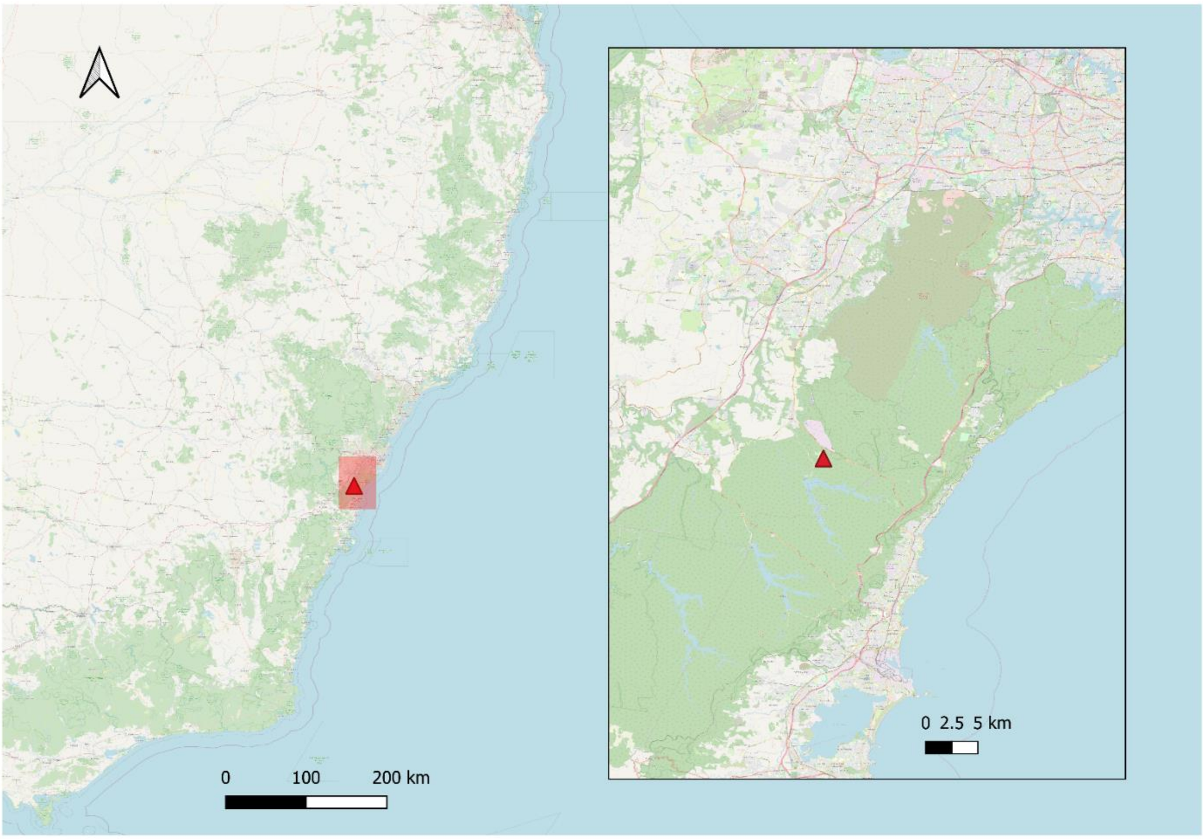

Figure 1 Location of the sampling site, to Sydney, NSW in the north. The sampling site had four different climate control containers for the instruments, as well as a soil sampling site around 50 meters northeast from the main sampling site and the High-Vol PM filter. "Map data copyrighted OpenStreetMap contributors and available from https://www.openstreetmap.org"

2.2 Instrumentation

The instruments deployed in the campaign are presented in **Table 1**. They included an air quality monitoring station owned and operated by the NSW Government Department of Climate Change, Energy, the Environment and Water (DCCEEW), located approximately 10 m away from the main sampling line for VOCs. This station included measurements of temperature, windspeed and direction, PM$_{10}$, PM$_{2.5}$, O$_3$, SO$_2$, NO$_x$, CO and visibility. Inlet heights on this station were between 4.5m to 5.6 m above ground level. All NSW air quality monitoring stations are accredited by the National Association of Testing Authorities (Australia), however it should be noted that these instruments are targeted at regulatory standards and are not research grade. In particular this means that measurements made close to the detection limits are likely to be inaccurate and should be interpreted as indicative measures rather than accurate quantitative measures of atmospheric concentrations.

VOCs were measured using a Proton Transfer Reaction Mass Spectrometer (Ionicon PTR-ToF-MS 4000) which operated with a mass range spanning m/z = 18-256. The drift tube was held at a temperature of 70° C, pressure at 2.60 mbar, and an electric field to molecular number density ratio of 120 Td. The instrument was housed in a separate climate-controlled unit. Samples were drawn from an inlet on a 10 m mast through a 20 m long PTFE line using a bypass flow of 1.2-3 L min^{-1}. Calibrations were made on site using standardized cylinders containing 17 compounds including isoprene, monoterpenes, methyl vinyl ketone (MVK) & methacrolein (MACR), benzene, C_8-aromatics, and C_9-benzenes (Mouat et al., 2022). Mass spectra were integrated to produce data at 1 minute temporal resolution. Mole fractions were further averaged on a five-minute basis.

A suite of aerosol instruments were operated within in the Atmospheric Integrated Research Facility for Boundaries and Oxidative eXperiment (AIRBOX) container (Chen et al., 2019). Sample air was drawn from a common aerosol bypass inlet. The inlet was located 5 m above ground level for the following instruments:

1. A Ultrafine Condensation Particle Counter (UCPC TSI 3776) was used to measure condensation nuclei number concentration greater than 3 nm (CN_3) (TSI Incorporated, Shoreview, MI, USA). The instrument was operated at a sample flow rate of 300 mL min^{-1}. Measurements were recorded at 1 Hz temporal resolution.

2. A Scanning Mobility Particle Sizer (SMPS) was used to measure aerosol size distribution between 14 and 670 nm mobility diameter. Full scans of this size range were recorded every five minutes. The system consisted of an X-ray aerosol neutralizer and 3071 Long Electrostatic Classifier (TSI Incorporated, Shoreview, MI, USA) coupled to a 3772 CPC (TSI Incorporated, Shoreview, MI, USA). Sample was drawn from the same inlet as used by the UCPC.

3. Chemical composition of aerosols with diameter smaller than 1 µm (PM_1) were taken using a Time-of-Flight Aerosol Chemical Speciation Monitor (ACSM; Aerodyne Research Inc., Billerica, MA, USA). Mass concentrations of organics (Org), sulphate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), and chloride (Cl^-) in the aerosol fraction 40-1000 nm vacuum aerodynamic diameter range, referred to as PM_1, are reported. Measurements were taken at 10-minute resolution. Sample air was drawn from the aerosol inlet common to the CPC and SMPS and dried using a Nafion dryer to < 40% relative humidity before sampling.

Table 1: Instruments deployed during the COALA 2020 campaign and included in the present analysis.

Name of parameter	Instrument type
NO - NO_2 -NO_x	API T204
O_3	Ecotech 9810
PM_{10}	Thermo (TEOM) 1405A
$PM_{2.5}$	Thermo (BAM)5014i
SO_2	API T100
Black Carbon	Magee Scientific Aethalometer AE33
VOCs	PTR-ToF-MS (Ionicon)
CO - CO_2 - CH_4 - N_2O	FTIR in situ analyser
CN_3	TSI 3776
Particle number size distribution (14 nm to 660 nm)	SMPS
PM_1 mass composition	Tof-ACSM, Aerodyne
Wind Speed and Wind Direction	2D Ultrasonic anemometer
Temperature, Relative humidity	Vaisala HMP155
Photosynthetic active radiation (PAR)	

2.3 NPF Classification Method

The method proposed by Dal Maso et al. (2005) was used to classify the particle size distribution data. To apply the method the particle number density plots were made for each day during the campaign and the plots were visually inspected to identify if an event occurred on that day. A day of data was classified as an *event* if there was nucleation, and growth up to 25nm for at least two hours.

Once the events were classified, a logarithmic fit was applied to determine the geometric diameter of each mode. The data was manually divided in chunks of 10 minutes to visually inspect and determine the number of modes and the geometrical diameter range of each event (nucleation <25 nm, Aitken 25 nm -100 nm, accumulation >100 nm). Once those parameters were defined and included in the code, each event was divided in periods of time with similar distribution modes.

For illustration a hypothetical event lasting two hours was divided in two: one hour with simultaneous two particle modes (nucleation and Aitken) and then one hour with just one particle mode (Aitken). This is done to estimate an accurate geometrical particle diameter based on the number of modes. This avoided the problems of changes in the number of modes in the sample. Finally, the data was merged again to have a time series of number of particles predicted with the fit, number of modes predicted and geometrical particle diameter.

The algorithm works by providing the number of modes observed in the input dataset. Then it selects the provided model equation for each mode number and iterates over a hundred fits looking for the best fit. The Bayesian Information Criterion (BIC) was used to identify the best fit, looking for the lowest values. Once the best fit was selected, the total particle number estimated by the model was compared with the sample record for each sample to assure it was within a 5 % difference compared to the total particle number reported in the sample. The result was then visually checked looking for the geometrical diameter and how it compares to the distribution size plots from the raw aerosol distribution size data. Once the model was considered representative and accurate enough, the growth rate for each event was determined using a simple linear regression of the change in the geometrical diameter in time from nucleation to Aitken and eventually to accumulation mode.

3. Results and Discussion

3.1 Frequency of NPF Events

Of the 40 days included in the analysis, nine days didn’t have any data. Of the 31 days with data, 14 (45%) showed clear NPF events, nine (29%) were considered undefined, eight (26%) didn’t have enough data or were classified as a non-event The percentage of days with NPF is similar to those of other sites in forested areas in the Northern Hemisphere (Kalkavouras et al., 2020; Uusitalo et al., 2021). 45% of days with NPF events and 29% with undefined events implies a highly reactive atmosphere even in this rural area with some anthropogenic influence of mobile sources and occasionally coal-fired power plant in the Hunter Valley region.

An example of the time series of an NPF event is shown in Figure 2 for the event on 11 Feb 2020. The NPF commences at 8 am shortly before which a peak is observed in SO_2 and the H_2SO4 proxy (estimated using the rural proxy proposed by Dada et al. 2020). The shaded area in the plot highlights the growth period which is marked by an increase in mode diameter and condensation sink. Ozone also increases in this time. The increase in aerosol SO_4 and organics during this period shows the influence of this reaction chemistry on particles larger than 100 nm in the aerosol size distribution

3.2 Triggers for NPF Events

Of the fourteen days with NPF, five occurred during the night or early morning (before sunrise), and nine during the day. The time series of SO_2, NO_x, ozone, VOCs and the aerosol composition were used to identify which variables triggered and influenced the aerosol formation and growth. Of the fourteen event days of NPF, eight days include VOC data and nine days include aerosol composition data, noting that the composition data is not applicable to particles <100 nm and only three events led to accumulation sized particles (diameter >100 nm).

From the daily time series of all available variables over the 14 days of NPF events, it is evident that SO_2 frequently triggers or at least influences the particle formation. However, the trigger for nighttime events is less clear. To group the common factors influencing NPFs for daytime and night-time events, a comparison of the growth rate was used to determine whether the rates were similar during the day and during the night.

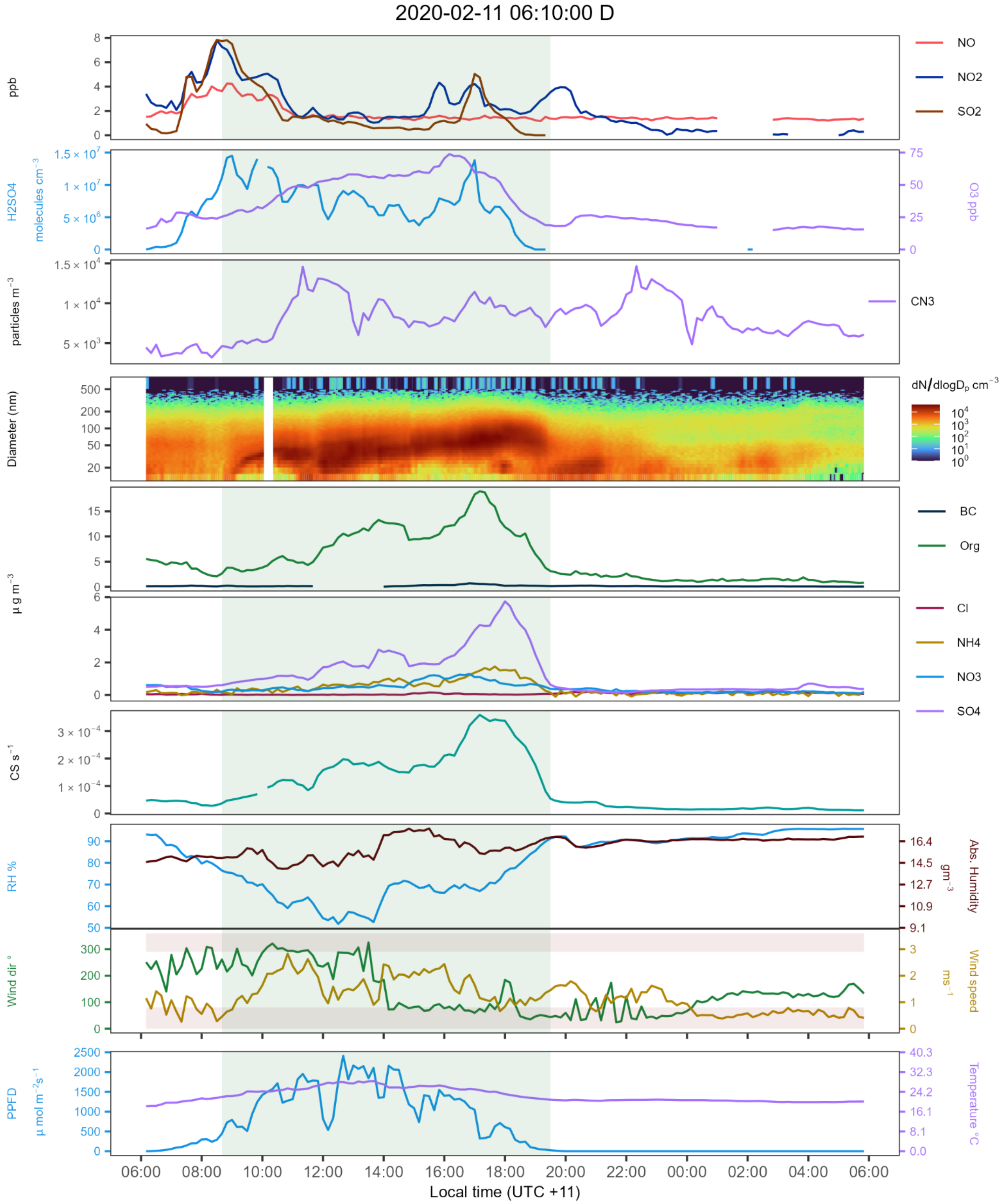

Figure 2 Time series for all selected variables during the NPF event during 2020-02-11. NO = Nitric oxide, NO2 = Nitrogen dioxide, SO2 = Sulphur dioxide, H2SO4 = Sulphuric acid, O3 = Ozone, CN3 = Condensation Nuclei >3nm, CN3-CN14 = difference of CN3 minus the sum of

all channels from the SMPS data. BC = Black carbon. Org = Organic mass fraction, NH4 = Ammonium mass fraction, NO3 = Nitrates mass fraction, SO4 = Sulphates mass fraction, Cl = Chloride mass fraction. CS = condensation sink. PFFD = Photosynthetic Photon Flux Density. VOCs mole fractions were not available during this specific event. Note how the fraction of organics, sulphates and ammonium increase with a positive correlation, dominating over the nitrate and chloride fractions until the end of the event. The light green area marks the NPF, and growth period mentioned in the analysis. the brown shade areas in the wind panel highlight areas where the wind comes from the nearby roads. Note that the NO values are close to detection limit and look biased high and hence should be interpreted as an indicative rather than accurate quantitative measure of atmospheric concentration.

3.3 Particle Growth Rates during daytime and nighttime events

The estimated growth rate is presented in Figure 3. Only four of the nine events during daytime (upper panel of Figure 3) had a representative Pearson coefficient (R>0.6), the remaining five events did not have a stable linear growth and are not shown in the plot. The events which showed unstable growth patterns suggests a highly variable condensation source, possibly resulting from changing H_2SO_4 concentrations. This is complicated further by changing wind directions.

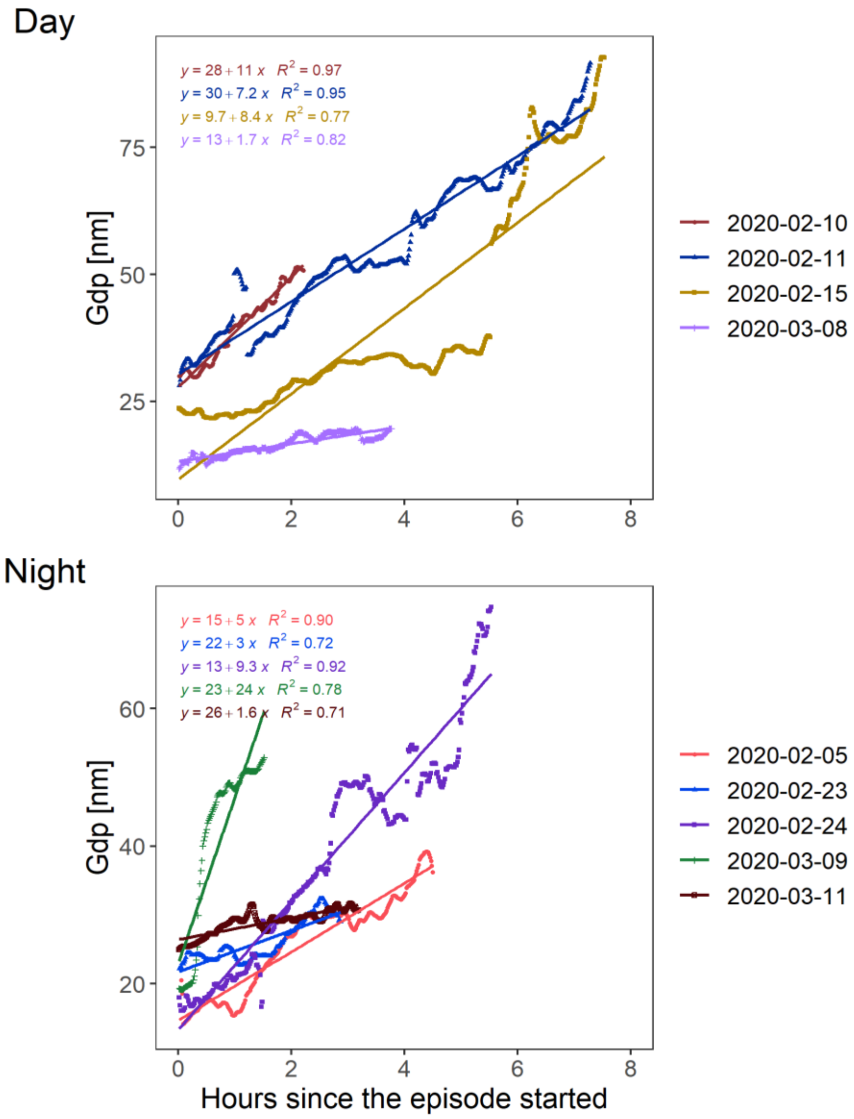

Figure 3: Geometric particle diameter evolution in each event where the logarithmic fit converged. The top panel presents the daytime data (only four events converged to a statistically significant model). The bottom panel presents the nighttime events.

Some events highlight how the dynamic nature of daytime concentrations of O_2, NO_2 and O_3 complicate the analysis (see figure S1). Nonetheless, these events provide insight into the factors that may drive the growth and particle formation and so were included in all the analysis. Event 2020-02-15 in Figure 3 is an example of how the geometric particle diameter can change when there is rapid growth. The first part of the regression shows slower growth rate. After the 6th hour of slow growth, the rate increases substantially.

In contrast to the daytime events, all the night-time events were stable enough to determine the event growth rate. The growth rate varied considerably between events (see lower panel of Figure 3) and most likely reflects differences in the factors driving the particle formation between these episodes. The differences in the growth rate might be directly affected by the main oxidation pathways available at the time of the reaction. These reaction pathways might include isoprene oxidation by nitrate radical (NO_3) oxidation path during the night (Wu et al., 2021b), monoterpene ozonolysis and condensation over previously formed clusters(Liu et al., 2023; Wang et al., 2023), or oxygenated VOCs (OVOCs) brought to the site and condensed on formed seeds or starting nucleation(Bianchi et al., 2019; Higgins et al., 2022). Some of these processes were observed during the campaign and will be further explored on the nighttime events section.

3.4 Daytime NPF Events

From the timeseries analysis of all daytime events (see Figure 2, 5-8 and supplementary figures S1-S5), four key points were identified for NPF in the area:

1. SO_2 arriving at the site appears to trigger nucleation and growth events.
2. VOC availability (monoterpenes and isoprene) enhances nucleation and growth.
3. The hours with high VOCs concentrations and higher oxidation capacity in the atmosphere (OH concentrations are assumed to be higher during the hours with higher PAR) have higher particle number concentrations and generally guaranteed growth up to the accumulation mode.
4. Growth without the influence of SO_2 may occur but will do so at a slower rate.

During most of the daytime events SO_2 and NO_2 plumes impacted the site at some stage of each event.

On some occasions the SO_2 plume might last for a couple hours as shown in the first part of the event on Feb 11th 2020 (see Figure 2), whilst at other times there were multiple peaks of high SO_2 measured at the site as shown in several other events in the record (e.g. Figures S2, S3, S4). However, subsequent nucleation was observed on every occasion that SO_2 was observed above the detection limit at the site, growth occurred within 0 to 150 minutes after the SO_2 was first detected. The time window difference between events reflects the influence of conditions at the start of a particle growth event. To highlight this phenomenon a cross correlation between SO_2 and the aerosol mass of aerosol SO_4 time series obtained from the tof-ACSM and the measured particle number concentration (CN_3) was applied. Figure 4 shows the Pearson correlation between SO_2 and the CN_3 and aerosol SO_4 in a window period of four hours i.e. starting two hours before the nucleation commenced and ending after the first two hours of the event. This time window captures the SO_2 influence on the particle formation. Each line/point shows the correlations at 0, 30, 60, 90, 120 and 150 minutes lagged for each daytime event. The dotted blue lines show where the lagged correlation is significant at ($|r| > 0.5$).

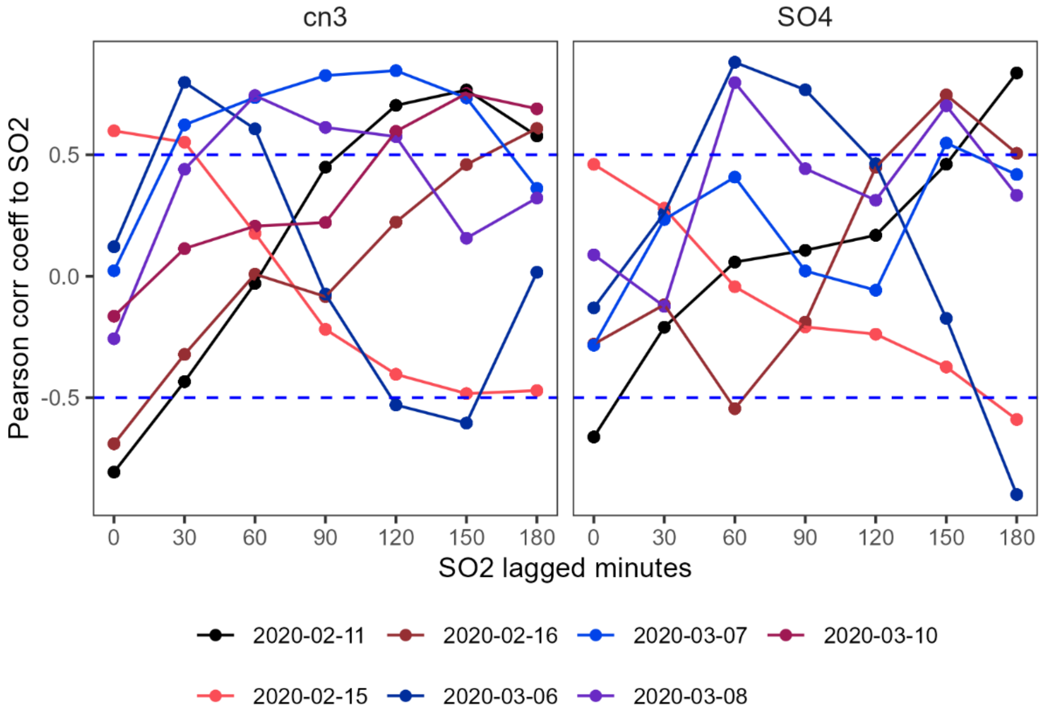

Figure 4 : Pearson correlation values obtained from the cross correlation between SO_2 and CN_3 and SO_4 mass. The dashed lines represent the 0.5 threshold as a reference to identify significant correlations. Events on Feb 10th and March 11th did not follow this pattern and were removed from the plot.

To interpret Figure 4, we can use the event on February 11th (black line) as an example. Here the correlation between SO_2 and CN_3 becomes significant (at $|r| > 0.5$) if the SO_2 time series is lagged 120 minutes with respect to the aerosol data; and the correlation between SO_2 and aerosol SO_4 becomes significant after 3 hours. This means that if we move the SO_2 time series two hours forward it will be better correlated with the particle number concentration, accounting for the reaction time of SO_2 to produce H_2SO_4 and enhance/trigger the particle formation under the conditions in the atmosphere at the time. Usually, the SO_2 correlation with aerosol SO_4 needs a longer lag time to be significant. This is a potential indication of the order in which the chemical reactions happen. First, we will see oxidation of the SO_2 to H_2SO_4, then nucleation, and finally growth in mass when there is condensation or coagulation near CCN sizes. Using time series analysis as shown here can provide more evidence when the chemical mechanisms are known but observations of other variables are not available.

A similar result is observed for other events at different lagged times. The difference in the time necessary to achieve a significant correlation between SO_2 and the particle number seems to be related to the quantity of VOCs available when the SO_2 plume arrives at the site This matches our understanding of the process from nucleation to particle growth, at early hours we observed monoterpenes levels that are sufficient to promote nucleation through ozonolysis and HOM formation (Iyer et al., 2021; Wang et al., 2023) but potentially not enough to promote growth to larger sizes as it seems most of the particle growth observed could be related to isoprene levels. Later with higher temperatures, emissions of isoprene increase so that

ELVOC or other OVOCs are potentially produced that contribute to growth in particle mass reflected in the increase of the sulphate fraction observed in the ACSM. Events on February 15th, March 06th and March 07th showed highest correlations in the first 30 minutes of lagging the SO_2 data. Common to all these events was a relatively high isoprene mole fraction and enhanced levels of monoterpenes (~1 ppb) in the hour before SO_2 being detected at the site (see Figure S1, S2 and S4) or in the circumstances where VOC data were not available, conditions where isoprene and monoterpenes mole fractions were assumed to be high by associated with weather conditions (i.e. high PAR and temperature see Figure S5). The high levels of monoterpenes and subsequent ozonolysis could be driving the particle formation at these times. In this period H2SO4 is available to drive the nucleation and the HOM proxy (monoterpenes*ozone (e.g.: Zhang et al., 2024)) at it’s highest values during the day potentially enhancing the effect.

The event on March 8th also met this condition (see Figure 5), although the event showed a relatively low growth rate. The isoprene and MACR + MVK concentrations in this event highlight the possibility of isoprene suppression (Heinritzi et al., 2020). The higher isoprene levels after 12:00, along with the increase of MACR +MVK indicate isoprene oxidation through OH. This is the first step in the reaction chain to produce C15 dimers. Although the CN_3 data is not complete for the event, it is clear there was a decline of the number of particles in the smaller sizes at the same time as the oxidation products of isoprene increase. This is also supported using the HOM formation proxy (monoterpenes*ozone), where at the times with higher particle numbers, the HOMS proxy is higher and then when the particle number decreases the HOM proxy does it too while MACR+MVK products increase suggesting a change to a more isoprene driven chemistry. The organic and sulphate fraction increase as well as the condensation sink suggesting larger particle formation and conditions that favour condensation and coagulation instead of nucleation.

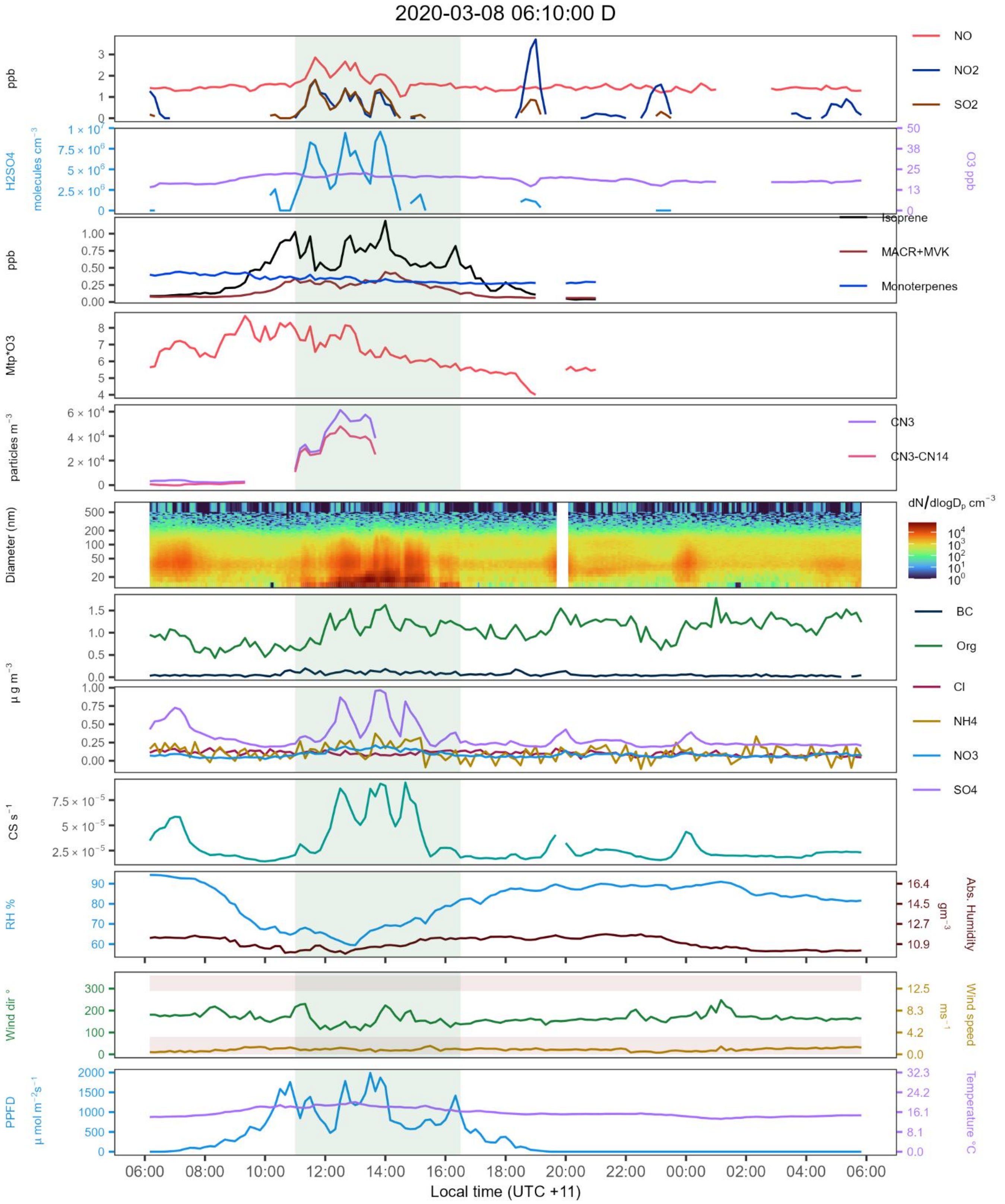

Figure 5 Time series of all selected variables during the NPF event during 2020-03-08. NO = Nitric oxide, NO2 = Nitrogen dioxide, SO2 = Sulphur dioxide, H2SO4 = Sulphuric acid, O3 = Ozone, MACR+MVK = isoprene ox. products methacrolein and methyl-vinyl-ketone, CN3 =

*Condensation Nuclei >3nm, CN3-CN14 = difference of CN3 minus the sum of all channels from the SMPS data. BC = Black carbon. Org = Organic mass fraction, NH4 = Ammonium mass fraction, NO3 = Nitrates mass fraction, SO4 = Sulphates mass fraction, Cl = Chloride mass fraction. CS = condensation sink. Mtp*ozone = HOM proxy product monoterpenes and ozone [ppb*ppb].*

The Feb 11th and Feb 16th events had similar arrival times for the SO_2 pollution (8:00 to 9:00) although the photochemistry was not fully active yet (see H_2SO_4), monoterpenes levels were consistently high during all the campaign (~0.4 ppb based on the days with data), enough to promote nucleation. This presumption is supported by looking at the event on February 16th (see Figure S3). In this event, a first peak of SO_2 at 8:00 started nucleation but then condensation or coagulation dominated favouring growth. The CN_3 -CN_{14} data show that after that initial nucleation period the particle number is dominated by the >14nm fraction. Multiple SO_2 plumes came to site producing higher ratios of H_2SO_4, but it promoted growth to larger particles sizes particularly on the sulphates fraction that correlates with the SO_2 peaks. In the evening there were a couple of small particle bursts that were quickly coagulated on larger size particles.

The event on March 10 (see Figure 6) shows high monoterpene concentrations that declined quickly just prior to the event being observed in the aerosol data. The aerosol growth phase is then observed to correlate with peaks in SO_2 and NO_X, as well as elevated levels of isoprene. Together, this suggests monoterpene ozonolysis initiated nucleation, followed by condensational growth via isoprene oxidation products.

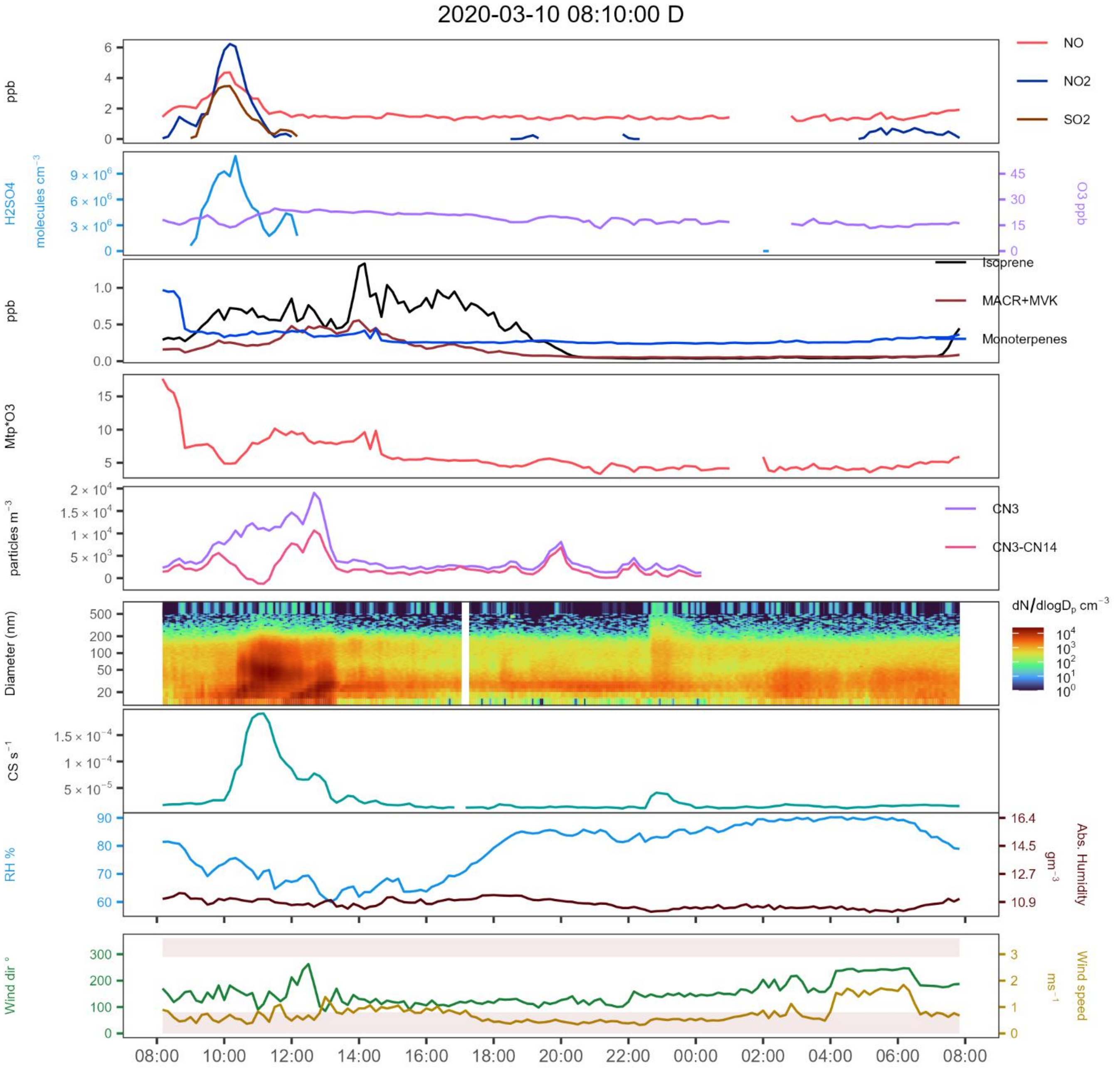

*Figure 6 Time series of all selected variables during the NPF event during 2020-03-10. The drop of CN_3 seem related to the lack of SO_2 after 11:00. NO = Nitric oxide, NO2 = Nitrogen dioxide, SO2 = Sulphur dioxide, H2SO4 = Sulphuric acid, O3 = Ozone, MACR+MVK = isoprene ox. products methacrolein and methyl-vinyl-ketone, CN3 = Condensation Nuclei >3nm, CN3-CN14 = difference of CN3 minus the sum of all channels from the SMPS data. CS = condensation sink. Mtp*ozone = HOM proxy product monoterpenes and ozone [ppb*ppb].*

For all date time events SO_2 and NO_2 are significantly correlated with a Pearson correlation of 0.78, suggesting a common source for both pollutants. The closest source of combustion products is the Appin Road located north of the sampling site. Given that the sampling site is away from other possible sources of SO_2 and NO_2 and the relatively low wind speeds during most of the campaign (see Figure S8), combustion from mobile sources is considered the most likely source of both compounds but there might be some influence of more distant coal-fired power stations. Another factor to contribute to this theory is that the SO_2 levels were higher during the day when most of the commuting takes place and leading to a higher vehicle density on the roads. The intermittent SO_2 and NO_2 peaks suggest the influence of mobile sources with poor emission control onboard. The effects of vehicles with poor emission control technologies on ambient concentrations of SO_2, NO_x , AVOCs and PM has been seen in different studies (Kari et al., 2019; Phillips et al., 2019; Smit et al., 2019) and the legislation controlling fuel standards and emissions is relatively lax in New South Wales (Paton-Walsh et al., 2019).

The atmospheric availability of monoterpenes to react and produce ULVOC or ELVOC with higher condensation potential to existing SO_4 aerosol is as important as the SO_2 presence and reaction, as seen in most of the events. Isoprene oxidation products also have a role in condensing on pre-existing nucleated aerosol. This has been previously reported by (Stangl et al., 2019), where chamber experiments showed that SO_2 presence can significantly enhance SOA formation from isoprene and monoterpene oxidation by ozone. Xu et al (2021b) reported that water and SO_2 availability will change the role of SO_2 in the particle formation process. With high SO_2 mole fractions, the SO_2 reaction path will favour reaction to peroxides instead of stable Criegee intermediates, thereby enhancing particle growth, particularly at relative humidity of greater than 45%; a condition present during most of the COALA-2020 campaign. This suggests that under high SO_2, isoprene and monoterpene availability, and high relative humidity conditions, particle formation and eventual growth is likely to occur.

Such an effect was observed in some of the events, for example the event on February 16th (Fig S3). This event saw high relative humidity throughout the event (well above the 45% threshold suggested by Xu et al.) as well as elevated gas-phase SO2. Unfortunately VOC data wasn’t available during this event, however the consistent diurnal profile of VOCs observed throughout the remaining dataset (Figure S5)can be extrapolated to this day, suggesting enhanced monoterpene and isoprene availability. Together, these prerequisites were met, and likely led to the observed aerosol growth event.

The conditions promoting sulphate formation in the event of Feb 16th are also present in multiple events. As the night approaches, BVOC emissions decrease with temperature, leaving all existing VOCs to oxidize. This provides the initiation for further oxidation of OVOCs into more oxidized species, which are more likely to condense on existing particles. In addition, with the temperature decreasing the relative humidity increases, making this the ideal condition for particle growth, particularly sulphates. However, once the VOCs are mostly consumed (assumed from diurnal cycles from the campaign to be around 22:00, see figure S5), there are insufficient VOCs to promote growth. This relationship is presented in Figure 7, where relative humidity and sulphate mass are positively correlated during the afternoon hours in the events which had multiple SO_2 peaks enhancing sulphate formation and particle growth.

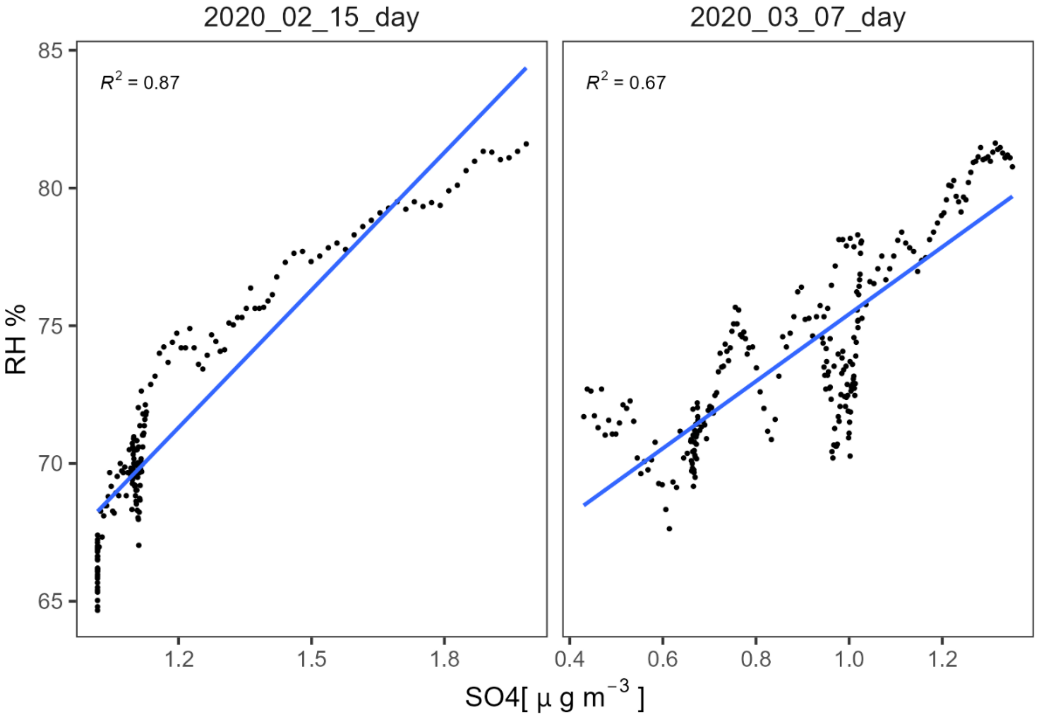

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Figure 7: Scatter plot of the events on Feb 15th and March 7th comparing total sulphate mass against relative humidity in the afternoon hours 389
(14:00 to 17:00). 390

When there is negligible SO_2 in the atmosphere but there are high enough VOC concentrations, there can be 391
particle growth when dimers C_{15}, produced by further OH oxidation of isoprene products, condensate over 392
smaller particles (Heinritzi et al., 2020). Growth was observed during the first event on February 10th (see 393
daytime data in Figure 8) and may be related to the condensation of these dimers. The average concentration 394
of isoprene during the campaign in the morning is higher than 1 ppb, enough to initiate the reactions leading 395
to LVOCs favouring growth of preexisting particles. 396

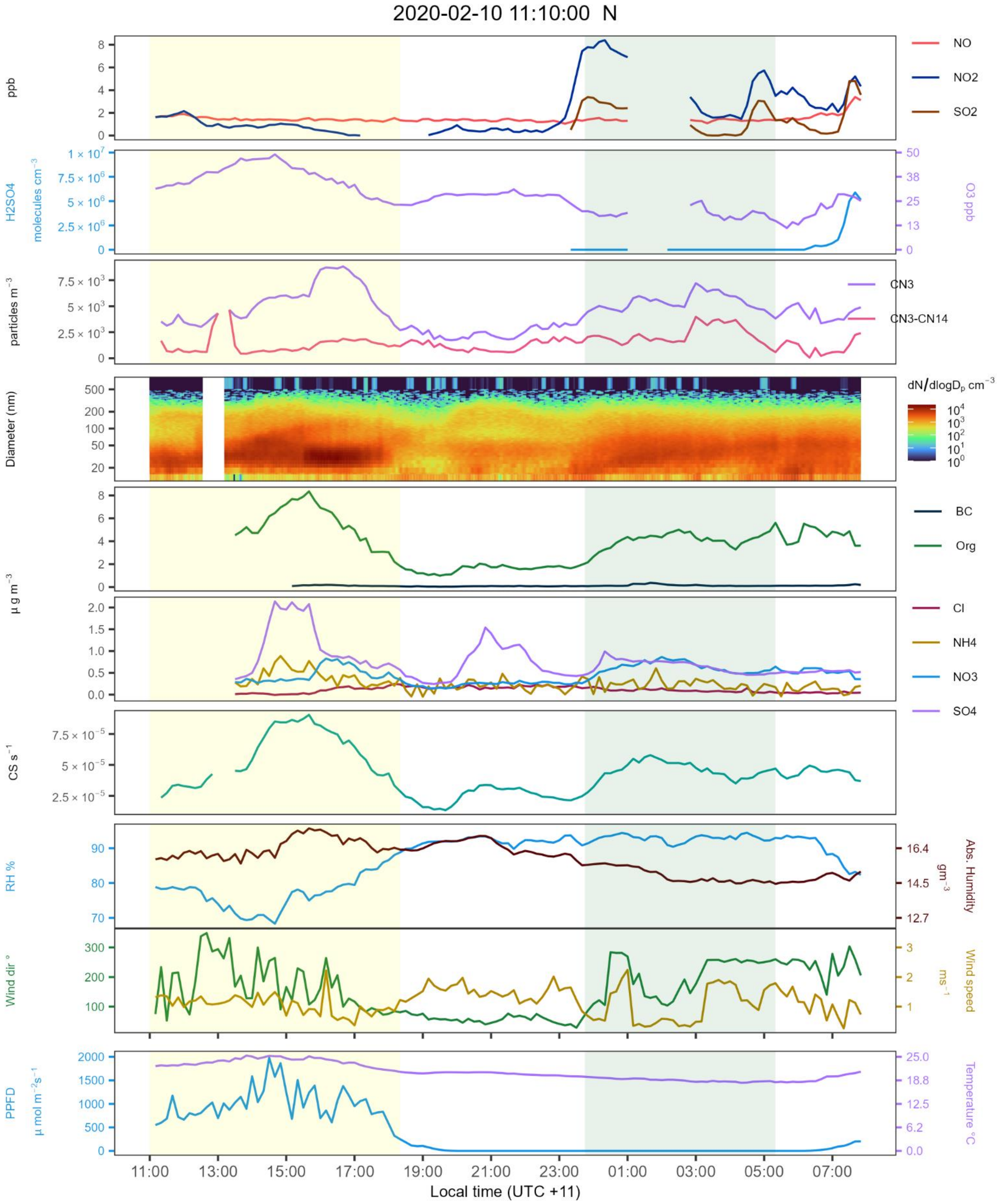

Figure 8: Time series for all selected variables during the NPF event during 2020-02-10. NO = Nitric oxide, NO2 = Nitrogen dioxide, SO2 = Sulphur dioxide, H2SO4 = Sulphuric acid, O3 = Ozone, CN3 = Condensation Nuclei >3nm, CN3-CN14 = difference of CN3 minus the sum of

all channels from the SMPS data. BC = Black carbon. Org = Organic mass fraction, NH4 = Ammonium mass fraction, NO3 = Nitrates mass fraction, SO4 = Sulphates mass fraction, Cl = Chloride mass fraction, CS = condensation sink. PFFD = Photosynthetic Photon Flux Density VOCs mole fractions were not available during this specific event. Note how there does not seem to be any significant SO_2 *or* NO_2 *pollution prior to the NPF start. At the same time of the particle growth there are enhancements in the organic, sulphate and ammonium mass fraction. There are two events in this plot. One in the morning with an unknown start and ending around 18:00, and the other at night. The light green area marks the night event, and the yellow highlight refers to the daytime event.*

The availability of monoterpenes increases the likelihood of NPF before or after SO_2 is available in the atmosphere. Although monoterpenes are quickly oxidized by OH resulting in relatively short lifetimes compared to isoprene (Atkinson, 2000; Atkinson and Arey, 2003), the ozone levels observed during the campaign are enough to promote ozonolysis and nucleation when there is no OH competing.. Australia experiences an isoprene dominated atmosphere (Emmerson et al., 2016; Ramirez-Gamboa et al., 2021) so the chemical balance in the atmosphere can rapidly change, particularly in the hotter seasons when more isoprene is emitted. Isoprene oxidises mainly through the OH pathway to more stable compounds; usually MACR and MVK are used as tracers to determine which path and under what conditions isoprene is oxidised. MACR is oxidised to heavier OVOCs that eventually condense and promote SOA formation in the larger sizes but these compounds also suppress NPF (Heinritzi et al., 2020; Link et al., 2021) as previously discussed in the event in Figure 5.

3.5 Night-time NPF Events

The main factors influencing the night-time events are ozone and NO_x pollution, however the data available for this study does not provide enough information to make a more definitive statement. Unfortunately, the NO_x instrument available in this study was not ideal for this type of measurement for several reasons: it is not designed to be sensitive to the low NO_x levels observed in rural areas; it is not capable of separating NO_x from NO_y; and it was set up to calibrate in the night hours between 1:00 and 2:00 every day. Nonetheless, during the night-time events the particle size distribution data and the CN_3 data showed particle formation and growth from nucleation to Aitken modes when there were considerable increases of NO_2 and simultaneous decreases in ozone. However, PM_1 aerosol mass from the ACSM showed a minimal increase at the same time (e.g. event during night of Feb 5th shown in Figure 9). This suggest that there are conditions to initiate the particle formation process but the conditions to increase the size/mass of the particles are not present at these times.

The night time nucleation observed in Figure 8 is related to an airmass change and might be the result of a combined effect of monoterpene ozonolysis and subsequent OH production after Crieege intermediates decay (Lester and Klippenstein, 2018). Once the OH is available it can produce H_2SO_4 and enhance the nucleation process. In this case, the increases in organic and sulphate mass shortly after the ozone depletion and the increase in CS indicate a growth in existing particles that is visible in the larger sizes in both particle numbers and mass. CN_3-CN_{14} data suggest that there was some nucleation sporadically happening after the event started, but these particles were rapidly coagulated together with pre-existing larger particles.

The event on Feb 5th may indicate a combination of different factors at play. First, the monoterpenes and ozone levels could be triggering nucleation as observed during the daytime events, but there is a slight increase in monoterpene concentrations potentially driven by slower wind speeds and less mixing volume at night due to the reduced boundary layer height. Secondly, isoprene is observed to be steadily decreasing during this period. There are no enhancements of MACR+MVK so we could speculate that isoprene may be oxidised by the nitrate radical pathway. This is supported by the slight increase in the Nitrates fraction observed with the ACSM around midnight. Later that day around 4:00 am there is a second burst of small particles that follow the same pattern of monoterpenes/ozone. The monoterpene ozonolysis is also seeing in other night events (see Fig S8-S10).

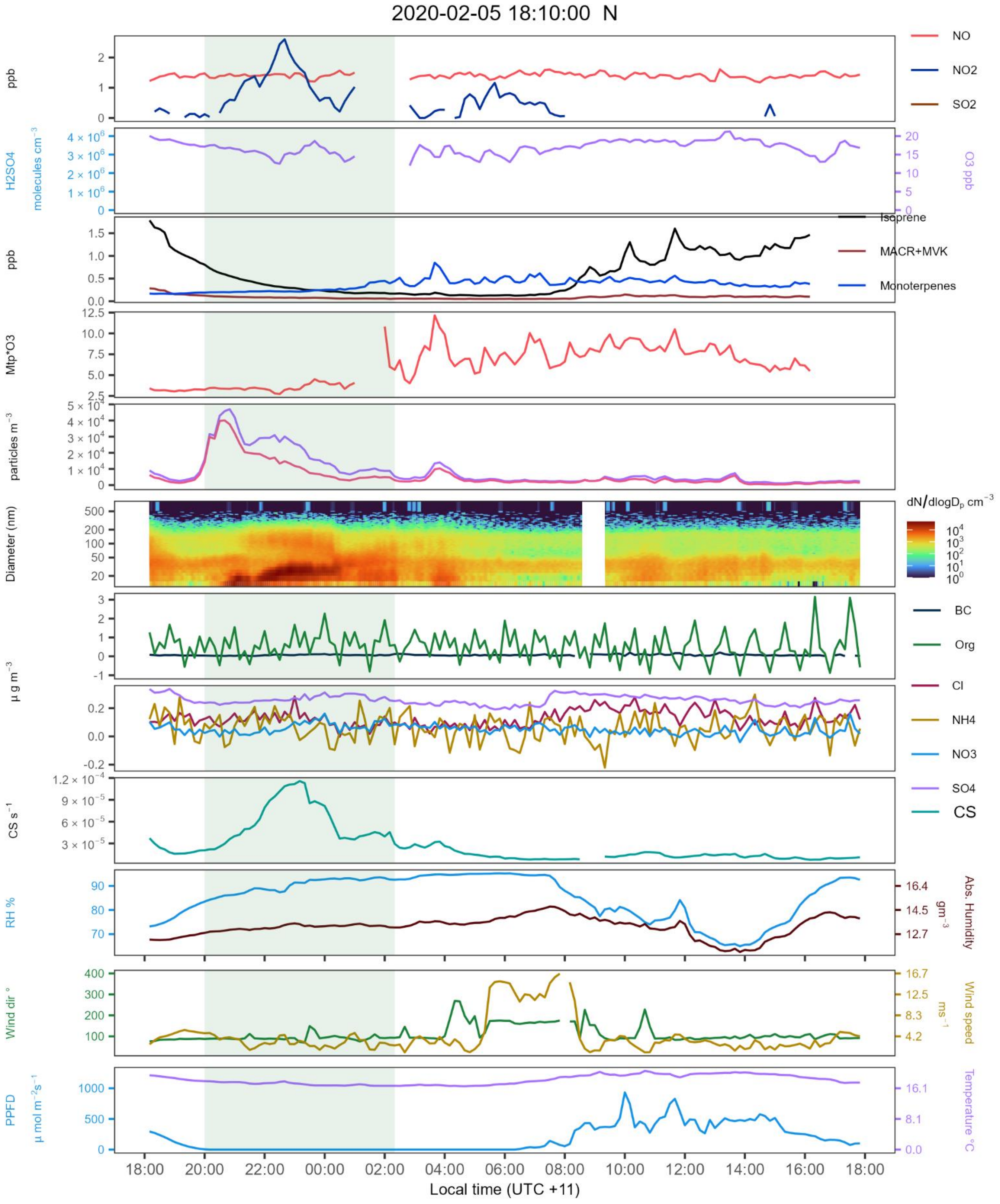

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Figure 9: Time series for all selected variables during the NPF event during 2020-02-05. NO = Nitric oxide, NO2 = Nitrogen dioxide, SO2 = Sulphur dioxide, H2SO4 = Sulphuric acid, O3 = Ozone, MACR+MVK = isoprene ox. products methacrolein and methyl-vinyl-ketone, CN3 = 446 447

*Condensation Nuclei >3nm, CN3-CN14 = difference of CN3 minus the sum of all channels from the SMPS data. BC = Black carbon. Org = Organic mass fraction, NH4 = Ammonium mass fraction, NO3 = Nitrates mass fraction, SO4 = Sulphates mass fraction, Cl = Chloride mass fraction. CS = condensation sink. Mtp*ozone = HOM proxy product monoterpenes and ozone [ppb*ppb]. Note how the particle number goes below 10000 after the growth reached Aitken mode (0:00). There is not a substantial increase in the aerosol mass when the particle number and geometrical particle diameter increase. The light green area marks the NPF and growth period mentioned in the analysis.*

3.6 Aerosol fraction: Day vs Night

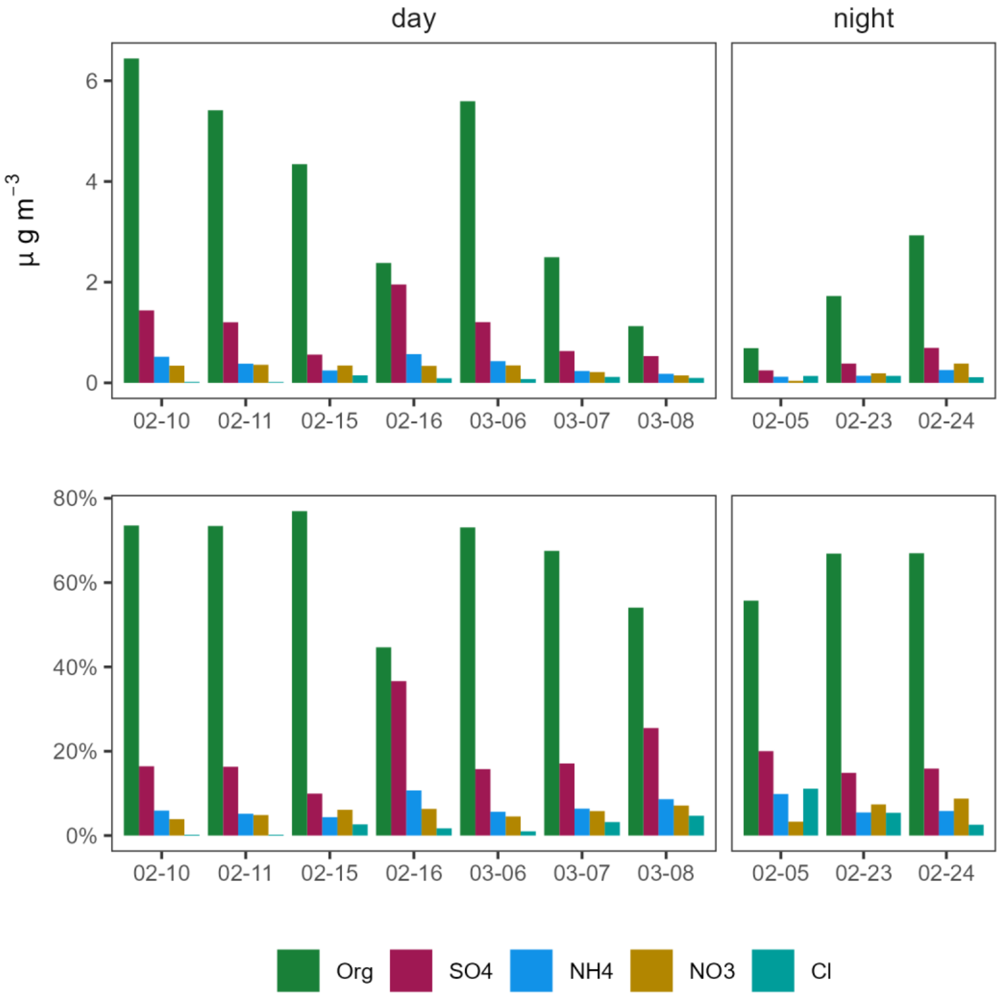

Figure 10: Average mass for each chemical group and event on the top panels. The bottom panel presents the percentage contribution to the mass of each of those fractions based on the average value presented above. Org = Organics; SO4 = sulphates; NH4 = ammonium; NO3 = nitrates and Cl = chlorides.

Figure 10 shows the mass fraction of the PM_1 aerosol mass as measured in the ACSM. Most of the daytime events show a similar mass fraction distribution. The organic fraction is the largest mass fraction followed by sulphates, ammonium, nitrates, and chlorides. We observed higher sulphate mass fractions in days with higher SO_2 availability such as the events on Feb 16th and March 8th, where the average sulphate mass fraction was larger or similar to the organic fraction (see Figure 10). These two events also display the highest proportion of ammonium during daytime events. The overall mass during night-time is much lower than during daytime, likely related to the lower concentrations of VOCs available during the night, resulting in growth not reaching sizes where it was detectable by the ACSM. Even with less total mass during the night, the contribution of each fraction is similar to the daytime events. The most notable difference between the mass fractions during day and nighttime NPF events is the higher fraction of chlorides during night-time attributed to a decrease in the aerosol formation capacity of the atmosphere at night that reduces the total organic, sulphate, nitrate and ammonia mass but does not impact chlorides as much.

Something to highlight is the higher fraction of ammonium compared to nitrates through most of the events. Regions with low NO_x have been previously characterized with higher ammonium fractions compared to nitrates (Du et al., 2015; Liu et al., 2022; Petit et al., 2015; Takami et al., 2005; Topping et al., 2004), whilst regions with higher NO_x concentrations favour nitrate formation (Hu et al., 2015; Parworth et al., 2015; Poulain et al., 2020; Schlag et al., 2016). The urban vs rural difference in relative mass composition is evident when comparing this study with the aerosol mass fractions observed in an urban site in Sydney (Keywood et al., 2016) in which high nitrate fractions were observed during most of the campaign.

4. Summary and Conclusions

Here we present aerosol concentration and composition data, VOCs and air pollutant concentrations collected during part of the COALA-2020 campaign including data from 5th Feb to 17th March at a rural site south of Sydney, Australia. This period followed the Black Summer fires after heavy rainfall cleared the smoke, offering insights into atmospheric processes under clean background conditions.

The atmosphere during the sampling period was classified as highly reactive with particle formation identified on more than 50% of the sampling days. Like previous studies, daytime NPF events coincided with the arrival of anthropogenic plumes at the site, suggesting their role in initiating particle formation. The positive relationship between isoprene concentrations and both PM_1 organic aerosol mass and CN_3 suggests a direct relationship between biogenic emissions and organic aerosol formation.

The change between gas to aerosol phase was indirectly analysed through the evaluation of the conditions leading to NPF events. This analysis showed how SO_2 plumes impacting the site drove NPF. The particle growth rate was dependent on available VOCs in the atmosphere and OH availability, also enhanced during periods with higher relative humidity and multiple intrusions of SO_2 and NO_x plumes producing particles larger than 100 nm.

Night-time events were attributed mainly to oxidation with ozone, and some potential growth through the isoprene/nitrate radical oxidation pathway. Although most of the night-time events showed the influence of monoterpene ozonolysis on NPF events, our data was limited and we acknowledge that other factors may have influenced nighttime NPF.

The COALA-2020 campaign highlights the significant role of biogenic emissions, particularly monoterpenes driving NPF and isoprene enhancing particle growth in Southeast Australia. These findings contribute to a better understanding of local atmospheric chemistry and its potential impact on regional air quality and climate. However, longer-term observations are necessary to capture the full picture of seasonal variations and non-fire related extreme events.

Supplementary Materials:

Author Contributions:

The experiment design was made by Clare Paton-Walsh (Murphy) and Melita Keywood.

The data collection was done by Jack Simmons, Travis Naylor, Paton-Walsh (Murphy), Asher Mouat, Melita Keywood, Ruhi Humpries, Malcolm Possell and Jhonathan Ramirez-Gamboa.

The data processing to convert mass spectra to concentration of VOCs was done by Asher Mouat under the direction and supervision of Jennifer Kaiser.

The data analysis was done by Jhonathan Ramirez-Gamboa

The paper was written by Jhonathan Ramirez-Gamboa and Clare Paton-Walsh (Murphy).

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Data Availability Statement:

Data is available at PANGEA via the following links:

- VOCs: https://doi.org/10.1594/PANGAEA.927277
- Aerosol size distributions: https://doi.org/10.1594/PANGAEA.928853
- Condensations nuclei > 3 nm in diameter: https://doi.org/10.1594/PANGAEA.925555
- Cloud condensation nuclei: https://doi.org/10.1594/PANGAEA.928925
- Green-house gases: https://doi.org/10.1594/PANGAEA.927313
- Air Quality data: https://doi.org/10.1594/PANGAEA.929001
- Meteorological data: https://doi.org/10.1594/PANGAEA.928929
- ACSM data: https://doi.org/10.1594/PANGAEA.973272

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Conflicts of Interest:

The authors declare no conflicts of interest.

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