Clean Air and Urban Landscapes Hub

National Environmental Science Programme

# CAUL Project 7, Milestone 7.1.16 Report on Air Quality and Smoke

Clean Air and Urban Landscapes Hub

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## About the Clean Air and Urban Landscapes Hub

The Clean Air and Urban Landscapes (CAUL) Hub is funded by the Australian Government's National Environmental Science Program. The remit of the CAUL Hub is to undertake "Research to support environmental quality in our urban areas". This includes research on air quality, urban greening, liveability and biodiversity, with a focus on practical implementation of research findings, public engagement and participation by Indigenous Australians. The CAUL Hub is a consortium of four universities: The University of Melbourne, RMIT University, the University of Western Australia and the University of Wollongong.

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# CAUL Project 7 Report for completion of Milestone 7.1.17:

# Air Quality and Smoke

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As part of CAUL project 7, researchers worked towards a better understanding of:

1. smoke composition from wildfires, prescribed burns and domestic wood heating
2. emissions from hazard reduction burns
3. emissions from domestic wood-heating and
4. improving estimates of population exposure to pollutants in smoke.

There was a particular focus on characterising the chemical composition of smoke using spectroscopic measurements techniques. Research from CAUL project 7 smoke research included detailed analyses of the composition of smoke to assess the accumulative effects of all the pollutants in different toxicological classes, so that total potential health impacts may be better understood in the context of other pollution sources. This work was published in the following two papers:

1. Desservettaz, M., F. Phillips, T. Naylor, O. Price, S. Samson, J. Kirkwood and C. Paton-Walsh (2019). "Air Quality Impacts of Smoke from Hazard Reduction Burns and Domestic Wood Heating in Western Sydney." Atmosphere **10**(9): 557.

2. MacSween, K., C. Paton-Walsh, C. Roulston, E.-A. Guérette, G. Edwards, F. Reisen, M. Desservettaz, M. Cameron, E. Young and D. Kubistin (2019). "Cumulative Firefighter Exposure to Multiple Toxins Emitted During Prescribed Burns in Australia." Exposure and Health: 1-13.

In 2020, due to COVID19 restrictions, some of the planned fieldwork could not take place. This included work in collaboration with the NSW OEH bushfires hub, working towards an understanding of the different emissions scenarios that result from cultural burning practices such as that undertaken by the Mudjingaalbaraga Firesticks Program. Nevertheless, the very widespread smoke pollution from the Black Summer bushfires resulted in smoke impacting the biogenics campaign site at Cataract where many atmospheric instruments had been deployed as part of the COALA-2020 (**C**haracterising **O**rganic and **A**erosol **L**oading over **A**ustralia) campaign (see milestone report 7.1.16). For this reason the milestones for 2020 were modified so that time could be devoted to studying these fires from the measurements that had already been obtained.

The results of this work have been presented at a number of (virtual) scientific conferences (see list below) and are being prepared for publication in the scientific literature (see Appendix 1).

**List of scientific presentations on smoke work in 2020:**

American Geophysical Union Fall Meeting 2020

*Biomass Burning Observations From the 2020 Wildfire Season in Eastern Australia*. Mouat, A. et al.

*The carbon cycle of southeast Australia during 2019/2020: Drought, fires and subsequent recovery.* B Byrne, J Liu, M Lee, Y Yin, KW Bowman, K Miyazaki, D Pollard, et al.

Atmospheric Composition and Chemistry Observations and Modelling Conference 2020
*The COALA-2020 Campaign: Characterising Organics and Aerosol Loading in Australia.* Paton-Walsh (Murphy), C. et al.

*Characterising the persistent smoke plume over SE Australian population centres during the 2019-2020 'Black Summer'* (poster). Simmons, J.B. et al.

Clean Air Society of Australia and New Zealand Biomass Burning Special Interest Group Workshop: The 2019-2020 bushfire season
*The chemical composition of Australian bushfire smoke: exposure.* Paton-Walsh (Murphy), C. et al.

Atmospheric Chemical Mechanism Conference, USA
*The chemical composition of Australian bushfire smoke: exposure.* Paton-Walsh (Murphy), C. et al.

# APPENDIX 1: Draft scientific paper on smoke measured during the COALA-2020 campaign

# Characterising the persistent, widespread smoke plume from the 2019-2020 Australian bushfires.

Jack B. Simmons, Clare Paton-Walsh (Murphy), Melita D. Keywood, Ruhi S. Humphries et al.

## Abstract

It is critical to understand the chemical composition of smoke generated from fires in the forests of southeastern Australian from a climatological and air quality standpoint. This study presents comprehensive greenhouse gas and aerosol measurements from the historically significant 2019-2020 Australian bushfire season.

## Introduction

Biomass burning (BB) is the largest contributor to fine carbonaceous aerosol globally, and second largest emission source of atmospheric trace gases (Akagi et al., 2011). A significant portion of total biomass burning emissions can be attributed to emissions from wildfires, which provide a climatically relevant source of black carbon aerosol and greenhouse gases to the global atmosphere (Liu et al., 2014). Australia is a region of global significance in the biomass burning field as fires occur frequently (van der Werf et al., 2006) and Australian emissions (expressed as TgC $year^{-1}$) are estimated to contribute 6-8% to the global BB total (van der Werf et al., 2006; Kasischke and Penner, 2004).

Biomass burning is estimated to contribute 53±14% to Australia's annual black carbon aerosol emissions (Qi and Wang, 2019). Savanna fires are the dominant contributor (83%) to biomass burning $CO_2$ emissions in Australia (Shi et al., 2015). This fire type occurs annually, confined to the north of the continent. Emission factors for trace gas and aerosol species from Australian dry season savanna fires have been determined (e.g. Desservettaz et al., 2017; Meyer, 2015). Forest fires along the east coast of the Australian continent also contribute to biomass burning in Australia, responsible for 13% of BB $CO_2$ emissions annually (Shi et al., 2015). These fires are more episodic in nature, with area burnt varying significantly from year to year (Australian Government Department of Industry Science Energy and Resources, 2020). The variability of fire weather is influenced by a number of inter-annual climatological processes, most notably the El Niño Southern Oscillation (ENSO) (Dowdy, 2018; Harris and Lucas, 2019).

The atmospheric impact of forest fire smoke in along the east coast of Australia is reasonably well documented. Smoke from large fires in 2013 in the Blue Mountains region of New South Wales was demonstrated to have had detrimental impacts on air quality as far away as Brisbane, Queensland as well in closer population centers such as Sydney and Wollongong, N.S.W. (Duc et al., 2018). Emission factors (EFs) for $CO_2$, CO, $N_2O$ and $CH_4$ were calculated for these fires using in-situ Fourier transform infra-red spectrometer (FTIR) measurements from Wollongong, N.S.W. (Rea et al., 2016). Fresh smoke from the firegrounds of a series of prescribed burns throughout N.S.W. and Victoria from 2010-2015 were measured by open path FTIR (Paton-Walsh et al., 2014), Selected Ion Flow Tube Mass Spectrometer (SIFT-MS) and grab samples (Guérette et al., 2018). Emission factors were calculated for greenhouse gases ($CO_2$: 1620 ±160 g $kg^{-1}$ fuel, CO: 120 ± 20 g $kg^{-1}$ fuel, $CH_4$: 3.6 ± 1.1 g $kg^{-1}$ fuel) and a large range of volatile organic compounds from these fires (Paton-Walsh et al., 2014; Guérette et al., 2018), and a dependence on Modified Combustion Efficiency (MCE: $CO_2$/CO+$CO_2$) was observed for some species (Guérette et al., 2018). Filter measurements of $PM_{2.5}$ from the fireground of four prescribed burns in temperate eucalypt forest in Victoria, southern Australia have been used to produce emission factors for this air quality pollutant (Reisen et al., 2018). The median $PM_{2.5}$ EF of 38.8 g $kg^{-1}$ fuel during smouldering combustion was greater than a factor of two bigger than the median EF measured during flaming combustion (16.9 g $kg^{-1}$ fuel). Measurements of trace species emitted by forest fire in south-eastern Australia have also been taken from satellite (Young and Paton-Walsh, 2011; Paton-Walsh et al., 2008) and aircraft platforms (Hurst et al., 1996).

The negative health implications of exposure to smoke components are well known e.g. (Lelieveld et al., 2015; Naeher et al., 2007; Pope III et al., 2002), specifically in the Australian context (Walter et al., 2020). Positive relationships between population exposure to forest fire smoke plumes and health metrics including adult asthma and chronic obstructive pulmonary disease hospital admissions (Morgan et al., 2010), non-accidental mortality (Johnston et al., 2011) and emergency ambulance dispatches (Salimi et al., 2017) have been observed in eastern Australian cities. Each of these studies use $PM_{10}$ measurements to define plume exposure. It should be noted that the majority of the negative health effect of exposure to bushfire smoke is attributable to the fine particulate pollution present in smoke (Johnston et al., 2012). However, a recent study has the contribution of gas phase toxics to smoke toxicity and that considering exposure to a range of air toxics is critical to determining health impacts of bushfire smoke in Australia (MacSween et al., 2019). Historically, particulate and CO pollution experienced in eastern Australia's population centres is attributable to smoke events (Duc et al., 2018; Paton-Walsh et al., 2019; Keywood et al., 2015).

The 2019-2020 Australian bushfire season was among the most significant in Australia's recent history. Large fires in temperate forest regions of South-eastern Australia were driven by historical dryness across the landscape coupled with meteorological conditions promoting wildfire (Nolan et al., 2020). It is estimated that 7.4 million hectares of temperate forest burnt from October 2019 to February 2020 (Australian Government Department of Industry Science Energy and Resources, 2020). Fires in temperate forests were widespread, stretching from near the N.S.W. -Queensland border to the Gippsland region of Victoria, a distance of over 1000 km. Thirty-four people were killed directly in fire events and thousands of buildings were destroyed. The fires were featured prominently in global news reports e.g. (News, 2020; Cave, 2020). Smoke caused exceedances of air quality monitoring criteria pollutants on an unprecedented scale at sites across Australia's east coast from Brisbane, Queensland (Queensland Government, 2020) to Melbourne, Victoria (Environmental Protection Authority Victoria, 2020). During the fire season (October 2019-February 2020), $PM_{2.5}$ concentrations exceeding the 95th percentile of historical daily mean values were observed by at least one monitoring station in Australia's eastern states on 94% of days (Borchers Arriagada et al., 2020). Borchers Arriagada et al. (2020) have attributed 417 (95% CI, 153–680) excess deaths to exposure to smoke from the 2019-2020 bushfire season along with thousands of hospital admissions. An early estimate of greenhouse gas emissions attributes 830 million tonnes of $CO_2$ equivalence from this fire season (Australian Government Department of Industry Science Energy and Resources, 2020), more than twice the emission from any fire season since 1990.

It should be noted that fire seasons similar to that which occurred in the 2019-2020 austral summer may become more frequent in the future. The frequency of fire weather in south-eastern Australia has increased in the past four decades likely due to the influence of anthropogenic climate change (Harris and Lucas, 2019). A trend is also observed in the onset of fire weather, with dangerous conditions occurring earlier in the year (Dowdy, 2018) lengthening the so-called 'fire season'. Forest fire frequency and intensity in the Australian southeast is predicted to increase into the future in a changing climate (Keywood et al., 2013).

The Characterizing Organics and Aerosol Loading over Australia (COALA-2020) campaign was a comprehensive atmospheric measurement campaign which occurred from January-March 2020 at Cataract Scout Park, close to Wollongong, N.S.W (Paton-Walsh et al., in prep.). While the primary objective of the campaign was to explore the relationship between biogenic VOC emissions and organic aerosol, the first three weeks of measurements were significantly impacted by bushfire smoke. Four concentrated smoke events were identified during this time, as well as "smoky background" period of aged, diluted smoke.

This study therefore aims to 1) present detailed measurements of both greenhouse gases and atmospheric particulates of smoke from the historically significant 2019-2020 Australian bushfire season as measured at COALA-2020 2) Present the general air quality characteristics of a 'smoky background' airmass that likely influenced a large portion of the surface atmosphere over the Australian east coast during the summer of 2019-2020., and 3) Provide a simple comparison of the Bureau of Meteorology AQFx smoke forecast model to observations. It is accompanied by a second study detailing coincident VOC measurements made at COALA-2020 on PTR-ToF-MS (Mouat et al., in prep).

## Methods

Cataract Scout Park (34°14’44“S, 150°49’26”E), at which the COALA campaign took place, is located twenty kilometres north-northwest of Wollongong on the east coast of New South Wales, Australia. The areas surrounding the site are heavily forested. Eucalypt species dominate the canopy. There are numerous longwall coal mine heads proximal to the measurement site. The Appin Colliery is approximately 1.5 kilometres to the north-northeast, and the West Cliff Colliery approximately 2.5 kilometres to the north. This site was selected for the COALA campaign as the primary objective of the campaign was to learn more about VOC emissions and secondary organic aerosol (SOA) formation from south-eastern Australian eucalypt forest. The Cataract site is distant enough from most anthropogenic emission sources (barring episodic influence from mine heads and proximal collieries) to provide a biogenic VOC background.

The COALA campaign was conducted from January 17th till March 23$^{rd}$, 2020, covering the second half of the Australian summer. Measurements presented in this study span from the beginning of the campaign until February 5$^{th}$, a period of 20 days. A widespread significant rain event occurred in NSW beginning on February 7$^{th}$, “cleaning” the local atmosphere of smoke and extinguishing most fires in the state (Bureau of Meteorology, 2020). Rain associated with this event leaked into a facility containing instrumentation for the campaign, forcing the shutdown of some measurements. Section 4.2 includes analysis of a longer period of measurements of N.S.W. criteria air quality pollutants, spanning December 20th, 2019 until March 24th, 2020. This analysis is possible as the air quality monitoring station measuring these species was able to be installed at the Cataract site early, before the bulk of the equipment.

During the analysed period, smoke from wildfires was the dominant influence on gaseous species and particulate variables measured.  Large, mature wildfires were burning in New South Wales throughout the analysed period, with many igniting in December 2019. The local weather was hot and sunny for the first two weeks of measurements, with most days experiencing maximum temperatures close to or above 30°C (hottest maximum: 43°C). The weather became cooler and cloudier during the last week of measurements presented with maximums closer to 20°C. Local winds were predominantly light (mean w/s 1.9 $ms^{-1}$) and variable, with a significant easterly contribution. Southerly and westerly winds were dominant during concentrated smoke events.

# Instrumentation

Detailed descriptions of all instruments used in COALA are provided in an overview publication (Paton-Walsh et al., in prep.). This publication focuses on greenhouse gas and particle-phase measurements of bushfire smoke. Instruments used heavily in the present analysis are described below. All measurements were averaged to five-minute time resolution for analysis.

### Fourier Transform InfraRed Spectrometer (FTIR)

A Spectronus FTIR (Griffith et al., 2012) was used to make continuous, high-precision measurements of $CO_2$, CO, $CH_4$, $N_2O$,  $^{\partial 13}C$ in $CO_2$ and $^{\partial 18}O$ in $CO_2$. This instrument produces GHG retrievals at two-minute resolution. It functioned near-continuously for the measurement period. Ambient air was drawn through approximately 7 m of ¼” Dekabon tubing from an inlet 4.7 m above ground level, before being dried by Nafion dryer and magnesium perchlorate before passing into the instrument. A calibration point was measured each day from a standard cylinder.

### Ultrafine Condensation Particle Counter

A TSI 3776 Ultrafine Condensation Particle Counter (CPC) 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 $min^{-1}$. Sample air was drawn from a common aerosol bypass inlet. The inlet was located 5 m above ground level. Measurements were recorded at 1 Hz temporal resolution.

### Scanning Mobility Particle Sizer (SMPS)

A SMPS was used during COALA 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.

### Multi Angle Absorption Photometer

The 5012 Multi-Angle Absorption Photometer (MAAP; Thermo-Fisher Scientific, Waltham, MA, USA) is used the measure the concentration of black carbon aerosol. Sample was drawn down a ½" inlet at 1000 $Lmin^{-1}$. Measurements of black carbon aerosol concentration were recorded at 1 Hz resolution. The inlet was located 4.8 m above ground level, and equipped with a $PM_{10}$ inlet cap.

### Air Quality Monitoring Station

An air quality monitoring station (AQMS) owned and operated by the NSW Government Department of Planning, Industry and Environment (DPIE) was installed at Cataract Scout Park for the COALA campaign. This station included measurements of temperature, wind speed, wind direction, $PM_{10}$, $PM_{2.5-}$, $O_3$, $SO_2$, $NO_x$, CO and visibility. NSW DPIE air quality stations are accredited by the National Association of Testing Authorities and use Standards Australia methods for ambient air quality measurements where possible (Department of Planning Industry and the Environment, 2020). Further details of specific measurement techniques are available (Department of Planning Industry and the Environment, 2020). Inlet heights for the station were as follows (meters above ground level): temperature: 4.5, wind: 5.6,  $PM_{10}$: 4.9, $PM_{2.5-}$: 4.9, gases: 4.7, visibility: 4.9.

# Modelling

Output from the Australian smoke forecast model, AQFx, was compared to the observations at the Cataract Site. This model was developed by the Bureau of Meteorology (BOM) and CSIRO in partnership with other organisations and is an implementation of the CSIRO Chemical Transport Model (CTM). It is forced by the BOM's ACCESS numerical meteorological forecast model.  It is operated by the BOM.  The AQFx model was run for the entre measurement period, and is capable of a resolution of 1.3 x 1.3 km. This fine resolution, coupled with the widespread scale of smoke plumes across SE Australia during the measurement period and a lack of immediate point sources near the measurement site, ensures it can be assumed that the model output from the grid cell containing the point measurement can be compared directly to measurements in this instance.

# Data processing and analysis techniques

Data processing was completed in the software 'R'.

# Results and Discussion

## Greenhouse gas and particulate measurements from COALA-2020

A timeseries of the CO record from COALA-2020 is presented in Figure 1. Four concentrated smoke events were identified in this record. CO was selected to identify plume events as the signal in this species was dominated by influence from bushfire smoke. The timing of the four smoke events is presented in Table 1. Smoke2 is associated with the highest recorded concentrations of CO, with concentrations above 1100 ppb observed. Enhancements above 500 ppb are observed in all four events. Smoke3 and Smoke4 are complex events, each appearing to consist of multiple plumes impacting the site. The mean 'dilute smoke' concentration of CO is 105 ppb. This is approximately 150% of the background expected at a site of this type during an unpolluted period.

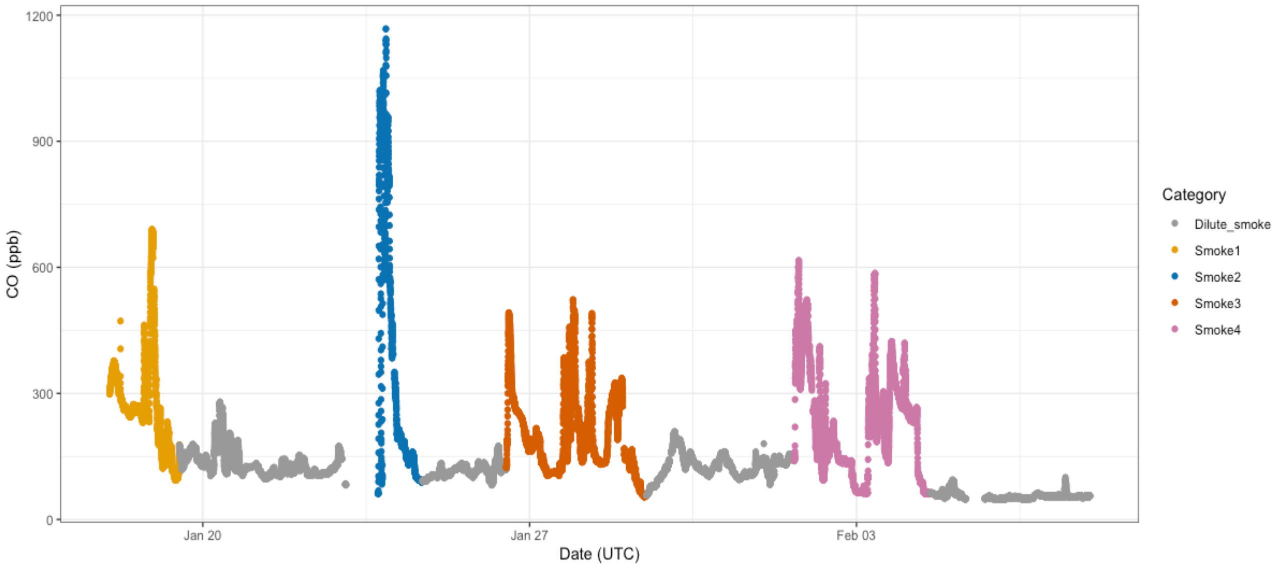


*Figure 1: Timeseries of carbon monoxide measurements from COALA-2020, limited to the smoke-influenced period of the campaign. Four concentrated smoke events have been identified from this record.*

*Table 1: Timing and duration of COALA-2020 concentrated smoke events.*

| Category | Start_UTC | End_UTC | Duration_hrs |
|---|---|---|---|
| **Smoke1** | 2020-01-18 00:00:00 | 2020-01-19 12:00:00 | 36.0 |
| **Smoke2** | 2020-01-23 18:00:00 | 2020-01-24 17:00:00 | 23.0 |
| **Smoke3** | 2020-01-26 12:00:00 | 2020-01-29 12:00:00 | 72.0 |
| **Smoke4A** | 2020-02-01 16:00:00 | 2020-02-03 05:30:00 | 33.5 |
| **Smoke4B** | 2020-02-03 05:30:00 | 2020-02-04 14:00:00 | 31.5 |

Enhancements in other sampled greenhouse gases are observed. A mean enhancement of 6.3 ppm above calculated background is observed in $CO_2$, whereas the mean $N_2O$ enhancement is 0.75 ppb compared to background. Methane is excluded from analysis at this point due to the confounding influence of local point sources- mine heads and cracks in the landscape releasing gas from subterranean stores. These point sources emit methane at very high concentrations (much higher than those found in bushfire smoke) in an episodic, irregular fashion. This makes the removal of such influence very difficult, and therefore the determination of an appropriate methane background highly uncertain.

Identified smoke events were also characterized by enhancements in other species. Black carbon aerosol which is expected to have a major source in bushfire smoke is highly correlated with CO. Maximum concentrations over 6 µg $m^{-3}$ were observed during the Smoke2. Aerosol size distributions were also influenced by smoke. Supplementary Figure S1 demonstrates a median particle size distribution observed during Smoke4 showing a significant mode in particle concentration at 100 nm. Smoke3 was only partially captured and therefore the plot is not representative of the whole event.

Interestingly, no enhancement is observed in the condensation nuclei > 3nm ( $CN_3$) record. Interestingly, higher concentrations of CN3 were observed after the smoke had cleared, possibly due to increased incidence of ultrafine particle formation and growth (Paton-Walsh, in prep.).

# Characterising air quality pollutants during the smoke period

The air quality monitoring station measuring N.S.W. criteria pollutants measured from December 20th, 2019 until March 24th, 2020. These measurements therefore captured many of the largest smoke plumes impacting the N.S.W. greater metropolitan region during the 2019-2020 fire season. Plotting timeseries of this period in Figure 2 for six criteria pollutants reveals significant enhancement during the smokey period (December 20$^{th}$, 2019 - February 5, 2020) especially in particulate pollutants.  It must be noted the concentrated smoke plumes detected with the full COALA-2020 instrument suite plotted in Fig. 1 are a small subset of the smoke plumes observed at the Cataract site throughout the summer.

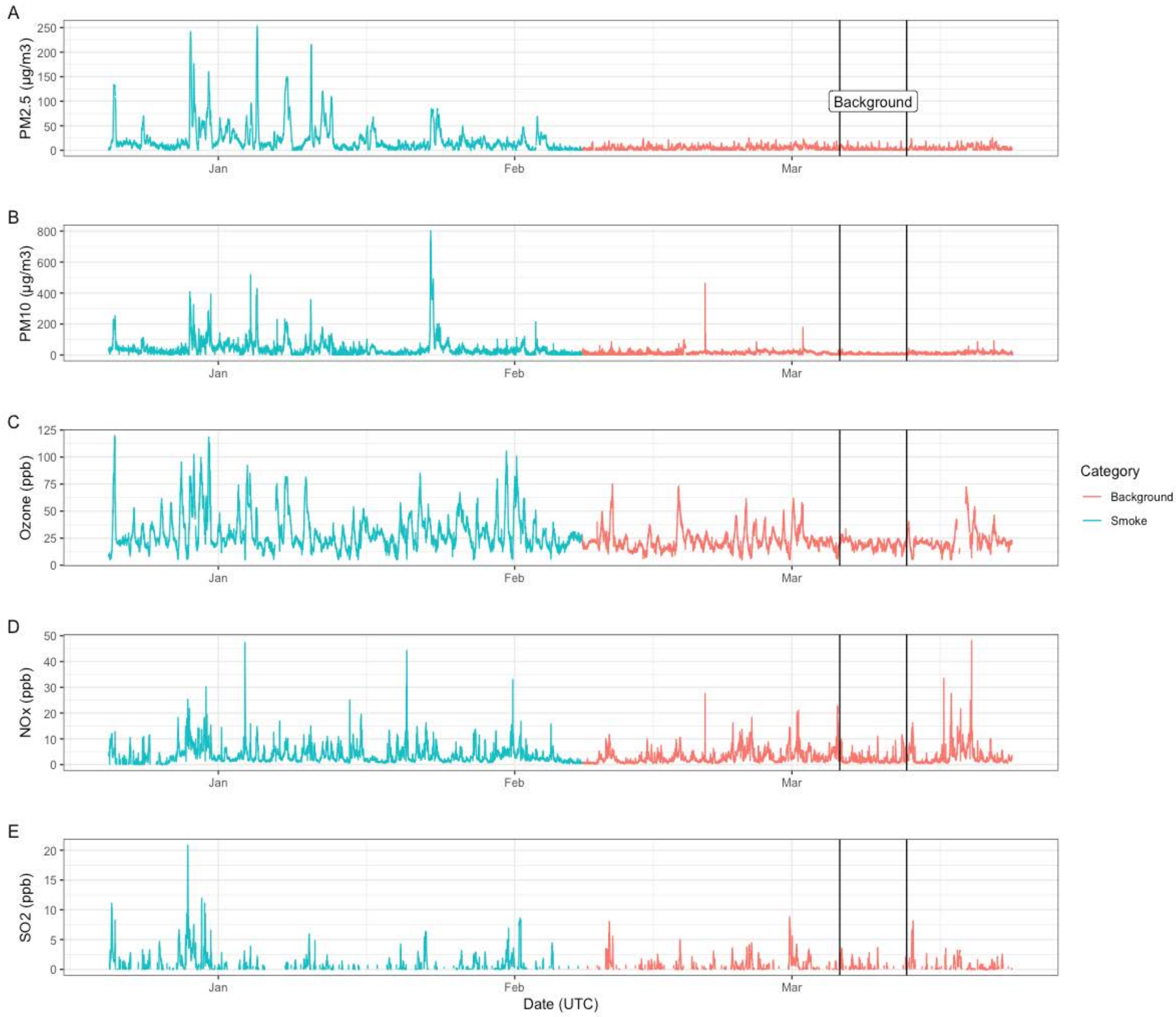


*Figure 2: Timeseries measurements of one-minute mean A) PM2.5, B) PM10, C) ozone D) $NO_x$ and E) sulfur dioxide for the duration of AQMS monitoring during COALA-2020. Timeseries are coloured to indicate measurements before and after the smokey period.*

Mean enhancements in absolute terms, and therefore inter-species enhancement ratios, can be calculated from this dataset. A background period was selected. This period, running from March 6, 2020 to March 13, 2020 had a mean temperature of 16°C, with a maximum of 23°C and a minimum of 13°C. The mean wind speed was $0.9ms^{-1}$. The nearby Holsworthy Aerodome weather station recorded 5 mL of rain on March 8th. This period was selected as it was cool and windy. Records of $NO_x$ and particulates suggest limited influence from proximal anthropogenic sources. Mean pollutant concentrations from this period are compared to the smokey period (December 20, 2019- February 5, 2020) in Table 2. A mean enhancement is calculated as the difference between the smokey period mean concentration and the mean background concentration.

*Table 2: Mean (standard deviation) air quality pollutant concentrations for the background and monitored smokey period (December 20, 2019- February 5, 2020) at Cataract Scout Park. The enhancement is the difference in means between the periods.*

| Category | PM2.5 (µg $m^{-3}$) | PM10 (µg $m^{-3}$) | Ozone (ppb) | NOx (ppb) | SO2 (ppb) |
|---|---|---|---|---|---|
| **Background** | 3.82 (3.45) | 8.61 (4.3) | 19.52 (3.63) | 2.04 (1.83) | 0.79 (0.78) |
| **Smoke Period** | 23.95 (30.06) | 45.89 (61.14) | 31.41 (17.71) | 3.4 (3.41) | 1.53 (2.03) |
| **Enhancement** | 20.13 | 37.28 | 11.88 | 1.93 | 0.75 |

All pollutants show significant enhancements during the smokey period. Large standard deviations in all smoke measurements indicate high levels of variability. This is expected considering the complex and variable mixture of fresh and aged smoke plumes passing over the site during the COALA-2020 campaign. It also indicates that the mean concentration is likely biased high, as the enhancement is unidirectional in most species (ozone excepted due to the effect of NOx titration). Enhancements are not surprising as wildfire smoke is a known and significant source of the reported air quality pollutants in Australia (Keywood et al., 2015; Duc et al., 2018; Rea et al., 2016). Comparing the observed values of these pollutants to the national concentration standard specified in the Australian National Environmental Protection (Ambient Air Quality) Measure (NEPM) demonstrates that bushfire smoke had a persistent and at times severe impact on air quality at the site. Legal limit concentrations defined by the NEPM (Australian Government, 2020), presented in Table 3, vary by pollutant and averaging period. A count of exceedances observed at the measurement site during the smokey period of COALA-2020 is presented in Table 4.

*Table 3: NEPM limit concentrations for air quality pollutants measured during COALA-2020. Categories with dashes indicate a lack of limit for the corresponding species and averaging time.*

| Averaging time | PM2.5 (µg $m^{-3}$) | PM10 (µg $m^{-3}$) | Ozone (ppb) | NOx (ppb) | SO2 (ppb) |
|---|---|---|---|---|---|
| **1 hour** | - | - | 100 | 120 | 200 |
| **4 hours** | - | - | 80 | - | - |
| **1 day** | 25 | 50 | - | - | 80 |

*Table 4: Exceedances of NEPM legal limit concentrations for air quality pollutants during the period December 20, 2019 - February 5, 2020 at Cataract.*

| Averaging time | PM2.5 | PM10 | Ozone | NOx | SO2 |
|---|---|---|---|---|---|
| **1 hour** | - | - | 5 | 0 | 0 |
| **4 hours** | - | - | 3 | - | - |
| **1 day** | 18 | 15 | - | - | 3 |

The air quality pollutants of most concern in N.S.W. are particulate pollutants $PM_{2.5}$ and $PM_{10}$, and ozone (Paton-Walsh et al., 2019). All of these pollutants were observed at concentrations greater than the NEPM limit on multiple occasions. Of forty-seven days during the AQMS-monitored period of COALA-2020, the daily NEPM limit was exceeded by $PM_{2.5}$ eighteen times- nearly 40% of total smokey days. In comparison during 2018 the Liverpool air quality monitoring station, the station with the most exceedances in greater Sydney metropolitan region, exceeded the daily limit concentration for $PM_{2.5}$ only eight times (Office of Environment and Heritage, 2019). Widespread observations of poor air quality around the N.S.W. during 2019 have been attributed to the influence of bushfire smoke, along with the preceding severe drought (Department of Planning, Industry and the Environment, 2020). Ozone pollution was less severe, with only five hourly exceedances and three four-hourly exceedances. These results are similar to results from a relatively polluted urban site during a period less effected by persistent bushfire smoke (Office of Environment and Heritage, 2019). The consideration of hot days, known to be associated with elevated ozone concentrations in Sydney, experienced during December and January at the COALA-2020 site (Utembe et al., 2018) is also necessary. Somewhat surprisingly, no hourly exceedances were recorded for $NO_x$ or $SO_2$ despite both these species being emitted as part of wildfire smoke. Three daily exceedances in $SO_2$ were observed.

## Determining enhancement ratios for air quality pollutants

Enhancement ratios (ER) can be calculated by examining the enhancement above background of one species compared to another reference species. This can be represented by the equation

$$ER_{\frac{a}{b}} = \frac{\delta[a]}{\delta[b]}$$

where $ER_{\frac{a}{b}}$ represents the enhancement ratio of species *a* to species *b*, $\delta[a]$ and $\delta[b]$ represent the enhancement above background for species *a* and *b* respectively (Desservettaz et al., 2017). This calculation assumes enhancements in both species can be attributed to a common source and requires a good definition of the background concentration of both species. As the COALA-2020 measurement site is removed from most large anthropogenic AQ pollutant sources, and the period of smoke influence is well defined, we can assume enhancements in species known to be emitted as part of bushfire smoke to be from a common source. A good knowledge of background is more problematic. In this case however, there is a record of significant length free from the influence of smoke that can be used as a background concentration. It is recognized that this approach introduces additional uncertainty into enhancement ratio estimates.

In this instance, CO is selected as the reference species. High precision CO measurements made using FTIR spectrometer began on January 18 and continued until March 15, 2020. Therefore the smoke period in the following analysis is limited to January 18- February 5, 2020. The background period as described in Section 4.2 is used to define mean background concentrations of measured species. Species with a significant diurnal pattern (e.g. $CO_2$) are corrected using a mean diurnal cycle calculated from the background period. The enhancements, as calculated in 4.2, for air quality pollutants for the period January 18 - February 5, 2020 are presented in Table 5.

*Table 5: Mean (standard deviation) air quality pollutant concentrations for the background (March 6 - March 13) and COALA-2020 smokey (January 18 - February 5) periods of COALA-2020. The enhancement is the difference in means between the periods.*

| **Category** | **CO (ppb)** | **PM2.5 (µg m$^{-3}$)** | **PM10 (µg m$^{-3}$)** | **Ozone (ppb)** | **NOx (ppb)** | **SO2 (ppb)** |
|---|---|---|---|---|---|---|
| **Background** | 70.52 | 3.82 | 8.61 | 19.52 | 2.04 | 0.79 |
| | (35.02) | (3.44) | (4.3) | (3.63) | (1.83) | (0.78) |
| **Smoke Period** | 175.21 | 15.34 | 39.22 | 31.61 | 3.4 | 1.33 |
| | (120.29) | (14.49) | (71.31) | (15.9) | (3.2) | (1.6) |
| **Enhancement** | 104.69 | 11.51 | 30.61 | 12.09 | 1.71 | 0.54 |

A large mean enhancement (105 ppb) in CO concentration is evident when comparing the background and smokey periods of COALA-2020. The enhancements for the period January 18 - February 5, 2020 are similar to those reported in Table 2 for the entire monitored period (December 20, 2019-February 5, 2020). The enhancement in the COALA-2020 (January 18- February 5) period is smaller for all species except ozone. $PM_{2.5}$ shows the greatest difference, with a mean enhancement during the entire monitoring period 43% higher than during COALA-2020. The mean enhancement in $PM_{10}$ is 18% higher, ozone is 2% lower, $NO_x$ is 11% higher and $SO_2$ is 28% higher in the entire monitoring period. This is expected, as it can be seen the most concentrated smoke events impacted the measurement site prior to January 18$^{th}$ (Fig. 2). The similarity in total enhancements shows that enhancement ratios calculated below may be applicable to fire events before the COALA-2020 campaign and represent a lower bound of smoke concentration from which enhancement ratios can be calculated.

Enhancement ratios were calculated using the method described above. Enhancement ratios for all air quality pollutants were calculated against CO and are presented in Table 6. The $PM_{2.5}$:CO enhancement ratio reported here fits in the range of literature reported values for wildfire emission ratios (~0.1-0.2 ug m$^{-3}$), which represent the ratio of pollutants to each other at the source. (Laing et al., 2017). Similarly the $NO_x$:CO ER reported here is close to the emission ratio reported in Akagi et al. (2011) for temperate forests.

*Table 6: Enhancement ratios against CO for air quality pollutants calculated during the COALA-2020 smokey period (January 18 – February 5, 2020).*

| **Ratio** | **Value** |
|---|---|
| **$PM_{2.5}$:CO (µg m$^{-3}$ per ppb)** | 0.110 |

| | |
|---|---|
| **$PM_{10}$:CO (μg $m^{-3}$ per ppb)** | 0.292 |
| **Ozone:CO (ppb per ppb)** | 0.115 |
| **$NO_x$:CO (ppb per ppb)** | 0.016 |
| **$SO_2$:CO (ppb per ppb)** | 0.005 |

Enhancement ratios of VOC species compared to CO were also examined and are presented in a companion paper (Mouat et al., in prep).

# Comparing model output to measurements

The BOM smoke forecast model AQFx output was retrieved for the Cataract measurement site, and modelled concentrations of air quality pollutants were compared to those observed during the smokey part of COALA-2020 (January 18th - February 5th, 2020). AQFx reports hourly pollutant concentrations and therefore comparisons were made to hourly mean observed values. Timeseries comparisons of CO and $PM_{2.5}$ are presented in Figure 3. The AQFx model captured CO concentrations well outside of the defined smoke events (Fig. 3A). The modelled timing of each smoke event is good, though the magnitudes of enhancements are too high during Smoke1, 3 and 4 and too low during Smoke2. The correlation coefficient for this comparison is 0.401.

Fig. 3B plots observed $PM_{2.5}$ concentrations against those simulated by AQFx. Again, the periods outside of defined smoke events seem to be reasonably well captured. There is some underestimation of particle concentrations during the maxima in Smoke2, the start of Smoke3 and the latter part of Smoke4. A large overestimation of $PM_{2.5}$ concentration is present on February 3rd. The correlation coefficient for this species is 0.224.

A dust event was observed during COALA-2020, identified by a large enhancement in $PM_{10}$ without a corresponding enhancement in CO, $PM_{2.5}$ or other species indicative of biomass burning or anthropogenic sources. This event occurred on January 23rd. 3C plots observed $PM_{10}$ concentration against the modelled $PM_{10}$ dust fraction, multiplied by a factor of 10. AQFx captures the timing of this event well. However, the magnitude is severely underestimated- by more than a factor of 10.

An enhancement is present in the AQFx records for both CO and $PM_{2.5}$ on January 22nd, evident in Fig. 3A and 3B. It appears AQFx predicts a smoke plume to impact the Cataract site. This plume is not evident in observations. This is the only significant example of a ‘false positive’ prediction in this dataset.

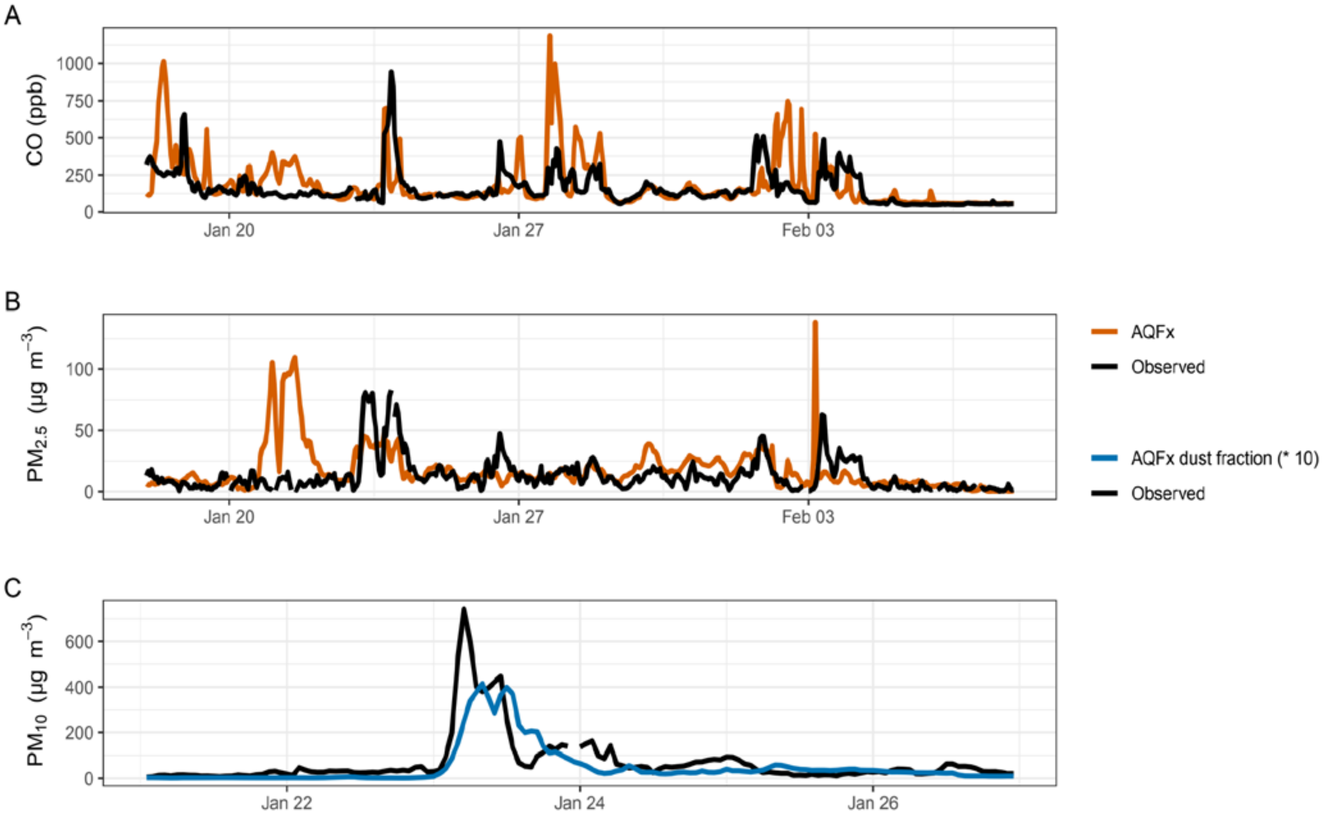


*Figure 3: Observations compared to AQFx simulations of A. CO and B. $PM_{2.5}$ during COALA-2020. C. plots the AQFx $PM_{10}$ dust fraction scaled by a factor of 10 against observations during a dust event sampled during the campaign.*

When evaluating the performance of AQFx in this context, it must be considered that the composition of the local atmosphere over the sampling site during this period was extremely complex, with dilute, recirculated smoke persistently impacting the site, coupled with fresher smoke from actively burning regions to the south and west. Considering this complexity, the AQFx model captures CO and $PM_{2.5}$ concentrations well for the majority of the smokey period of COALA-2020.

## Conclusion

The 2019-2020 Australian bushfire season was among the most severe in recorded history. Significant atmospheric chemistry and air quality impacts were observed across much of the east coast as a result of smoke plumes from wildfires, including in the majority of Australia's large population centres. This work presents greenhouse gas and atmospheric particulate measurements from a range of instruments, taken near Wollongong, N.S.W. during fire events. Four concentrated plume events are observed at the measurement site. We observe significantly enhanced concentrations in a wide range of species including CO, $CO_2$, black carbon aerosol and aerosol size distributions. We also determine the effect of biomass burning smoke during this period on air quality. Significant mean enhancements are also observed in ozone, $PM_{2.5}$ and $PM_{10}$, with health effects as a result of exposure to these pollutants reported elsewhere. Finally, we compare output from the AQFx smoke forecast model with observations and show the model performs well during periods of dilute smoke but may underestimate maximum concentrations during concentrated smoke events.

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# Supplementary

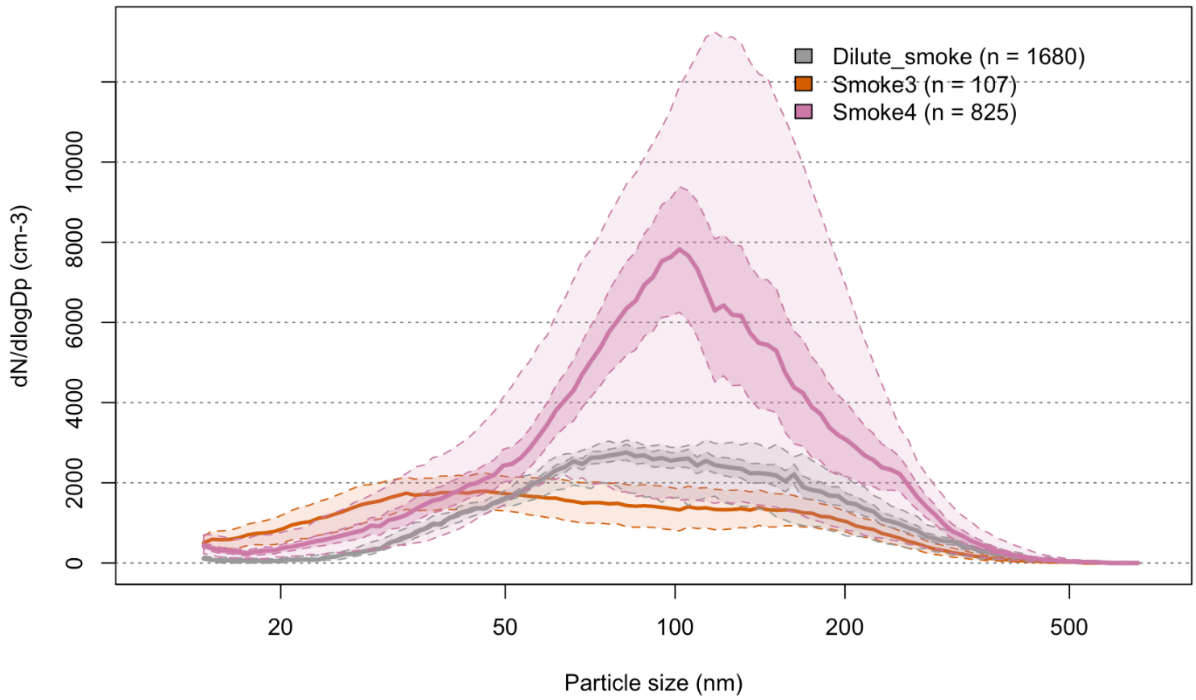


*Figure S1: Median particle size distributions for Smoke3, Smoke4 and the dilute smoke period. Note only the end of the Smoke4 event was captured, so the plot is not indicative of the entire event.*