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2019–20 Australian Bushfires and Anomalies in Carbon Monoxide Surface and Column Measurements

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Abstract: In Australia, bushfires are a natural part of the country's landscape and essential for the regeneration of plant species; however, the 2019–20 bushfires were unprecedented in their extent and intensity. This paper is focused on the 2019–20 Australian bushfires and the resulting surface and column atmospheric carbon monoxide (CO) anomalies around Wollongong. Column CO data from the ground-based Total Carbon Column Observing Network (TCCON) and Network for the Detection of Atmospheric Composition Change (NDACC) site in Wollongong are used together with surface in situ measurements. A systematic comparison was performed between the surface in situ and column measurements of CO to better understand whether column measurements can be used as an estimate of the surface concentrations. If so, satellite column measurements of CO could be used to estimate the exposure of humans to CO and other fire-related pollutants. We find that the enhancements in the column measurements are not always significantly evident in the corresponding surface measurements. Topographical features play a key role in determining the surface exposures from column abundance especially in a coastal city like Wollongong. The topography at Wollongong, combined with meteorological effects, potentially exacerbates differences in the column and surface. Hence, satellite column amounts are unlikely to provide an accurate reflection of exposure at the ground during major events like the 2019–2020 bushfires.

Keywords: bushfires; Australia; TCCON; column; carbon monoxide

1. Introduction

Fire is an essential global phenomenon that is often associated with terrestrial plants, and in a high spatially continuous biomass, like the temperate forest in Australia, large wildfires are driven by the availability of fuel that is sufficiently dry to burn, and therefore fuel dryness plays a key role in large wildfires in Southeastern Australia [1]. Vegetation fires have a major influence in shaping the ecosystem patterns and processes and regeneration of many plant species and can act as a natural clearing process that helps the ecosystem to grow and flourish. However, human activities have contributed to a changing climate and impacted fire regimes with the result that the fires have become more intense and severe in nature. For instance, fire weather conditions in southern Australia are becoming increasingly more dangerous, with earlier starts, increased frequency, and more extreme fire seasons [2], with modelling results indicating that climate change will make fire weather extremes more common in the coming decades [3,4].

In addition to being influenced by the changing composition of the atmosphere as greenhouse gas concentrations climb and warm the climate, fires emit large amounts of a wide range of trace gases and aerosols into the atmosphere [5]. Australian fires emit an average of around 130 Tg of carbon per year into the atmosphere, equivalent to 10–20% of all Southern Hemisphere and 6.7% of global fire emissions [6]. The magnitude

of these emissions, combined with the relative lack of industrial and fossil fuel sources in the Southern Hemisphere, implies that fires are a major contributor to atmospheric composition at the hemispheric scale [7]. The large variability in location and severity of biomass burning emissions from year to year means that fires are the largest contributor to short-term variability in tropospheric composition [8]. Although the majority of carbon emitted by Australian forest fires is in the form of carbon dioxide (CO_2), it is assumed that the net long term impact is zero as the CO_2 will be re-absorbed by the subsequent regrowth [5]. The long lifetime of CO_2 and high atmospheric background mean (monthly mean baseline CO_2 concentration measured at Cape Grim Baseline Air Pollution Station, Tasmania for December 2019 is 408.50 ppm [9]) indicates that it is not easy to track the impact of fires in observations of CO_2 concentrations. In contrast, CO has an annual average lifetime of 2 months and hence a relatively low atmospheric background concentration. It is most abundant in the Northern Hemisphere, with an average mixing ratio of approximately 150 ppb, while the Southern Hemisphere (SH) mixing ratio is around 50 ppb [10]. For these reasons, CO makes an excellent tracer of emissions from fires as it can be observed for several weeks after major fires at increased (or elevated) concentrations wherever the smoke plumes have been dispersed [11].

Due to the limited influence from anthropogenic emissions compared to Northern Hemisphere, emission sources of CO in the SH are dominated by biomass burning and chemical production from biogenic sources [12]. Hence, sites studied in the southern hemisphere show that CO concentrations tend to be affected by a balance of long-range transport, secondary organic production, and OH (hydroxyl radical) oxidation removal. It is evident from the previous studies that southern Africa and South America are the largest emitters of CO from vegetation fires in the SH; however, their interannual variability is much less than in Australia [6,13], making Australia an interesting place to study the atmospheric impacts of fires.

Surface in situ measurements of trace gases like CO provide highly accurate measurements of a point sample where the measurement is taken, however, they are highly sensitive to near-surface processes (such as mixing) in the planetary boundary layer [14–16]. On the other hand column measurements, based on the integrated vertical column, are less sensitive to surface processes and hence provide a robust measure for complementing the existing in situ network [17,18]. The Total Carbon Column Observing Network (TCCON) is a global network of ground-based Fourier transform spectrometers that remotely measure column abundances of trace gases, including CO, that absorb in near-infrared. TCCON is focused on providing long term, accurate and precise measurement to act as a primary validation data set for satellite measurements [19]. In contrast, on a global scale, satellite measurements are effective at providing spatial coverage of atmospheric composition but are less precise and have reduced temporal coverage compared to continuous ground-based measurements.

The 2019–20 fire season (also known as “Black Summer”, from August 2019–February 2020) in Australia was unprecedented in its extent, intensity, and number of fires [20]. According to the official records, New South Wales recorded a burned area of approximately 5.68 million hectares (7%) with 1.58 million hectares burning in Victoria (also 7%) [20]. Rainforests of Southeast Queensland were also widely impacted by the 2019–20 bushfires, with 97,200 hectares burned out of a total 2.4 million hectares, ~46% of it with high or very high severity [21]. The major reasons attributed to the occurrence of the unprecedented fires were the dryness in the fuel load due to the extreme drought conditions that prevailed during the spring and summer in south-eastern Australia [22]. The Black Summer fires injected approximately 0.9 Tg of smoke mass (sum of CO, acetonitrile (CH_3CN), H_2O and aerosols above the 380 K potential temperature, between 20°S and 82°S) into the stratosphere, which was responsible for a persistent warming of 1–2 K in the stratosphere for more than six months [23].

During the Black Summer fires, large populations in Australia were exposed to very high levels of smoke pollution over extended time periods. Several previous studies have

discussed the impact of these air toxins on human health. Inhalation of smoke can result in immediate physical and mental impairment [24–26] and long-term health effects such as an increased risk of cancer and a weakening of respiratory function [27–32]. A recent study by MacSween et al. [33] performed an analysis based on the prescribed burn data record in Australia to determine the cumulative exposure risk of firefighters to the mixtures of air toxins based on the current firefighter exposure regulations. The study found that the four major contributors to the exposure percent limit were respirable particulates, acrolein, formaldehyde and CO. Many trace gases and particulates that are emitted by fires are known to be highly correlated with CO in smoke plumes (e.g., [34]) and so measurements of CO have also been used to infer the likely concentrations of other toxic compounds in air. CO and respirable particulates are measured at a number of air quality stations in Australia but the observation sites are relatively sparse especially outside major metropolitan centres. It would therefore be advantageous to use data from satellite sensors such as MOPITT (Measurement of Pollution in the Troposphere) [35,36] or IASI (Infrared Atmospheric Sounding Interferometer) [37] to map the temporal and spatial distribution of total column amounts of CO and use this to estimate likely exposure levels on the ground. Such a process is predicated on the assumption that the total column amounts will be highly correlated with concentrations at the ground in these conditions. Several previous studies have shown that remote sensing is a useful tool for exploring the spatial variability of air pollution exposure in an urban area e.g., [38–44]. However, most of these studies are mainly focussed on short-lived aerosols and tropospheric trace gases such as nitrogen dioxide (NO₂) where short tropospheric lifetimes of ~1 day limit the effects of atmospheric transport; little work was done to test this assumption for a comparatively long-lived trace gas like CO.

Wollongong is an extratropical TCCON site located on the east coast of Australia, where the impact of the 2019–20 fires was significant. In addition to the column measurements made within TCCON, surface in-situ measurements of CO are also made at the same Wollongong site. Hence, this study focuses on comparing surface in situ and column measurements of CO to better understand whether column measurements can be used as an estimate of the surface concentrations. If so, satellite column measurements of CO could be used to estimate the exposure of humans to CO and other fire-related pollutants. Figure 1 presents the retrieved CO total column from MOPITT [45] during the peak bushfire episode in New South Wales. It clearly indicates the very high total column CO abundance ($2.30\text{--}4.96 \times 10^{18}$ molecules cm^{−2}) over Southeast Australia. High values of total column CO over Northern Hemisphere Africa can be attributed to the Northern Hemisphere biomass burning over the savanna south of the Sahara Desert and in the tropical rain forests, which often happens during December–April [11] while the high total column CO over Northeast China is mainly attributed to the anthropogenic emissions.

In this manuscript, we present a multi-year time-series of CO total column and surface observations from Wollongong, dating from 1997 and 2011, respectively, until present. We analyse these datasets in order to answer the following questions:

1. how anomalous were the black summer fires in the terms of the CO surface concentrations and column amounts at Wollongong?
2. how well do enhancements in total column amounts of CO at Wollongong match the enhancements in surface in situ CO measurements? (Here, we use the word ‘enhancement’ to mean the amount that CO is increased above the defined background)

We then discuss the implications of our findings for studies aiming to use satellite data of atmospheric total columns to estimate exposure levels at ground level.

Figure 1. MOPITT total column CO (version 7) data product from 20 December 2019 to 31 January 2020, (generated in: <https://www.acom.ucar.edu/mopitt/webview/plot-options.shtml>, accessed on 6 May 2021)

2. Methods

2.1. Site Description

The Wollongong measurement site is located at the University of Wollongong (UOW) (34.406° S, 150.879° E, 30 m above sea level) and is operated by the Centre for Atmospheric Chemistry (Figure 2). A detailed description of the site, meteorology and different possible pollution sources is provided by Buchholz et al. [46]. Briefly, Wollongong is a coastal city widely impacted by both local urban emissions from Wollongong city and the transported plume from Sydney (the most populous city in Australia, 80 km to the north of UOW). Since Wollongong is a narrow coastal strip between the 500 m high Illawarra escarpment and the sea, this site is continuously impacted by sea breezes, which create an inversion layer and often help in the trapping of atmospheric pollutants near to the ground [47].

Ambient CO measurements from two nearby air quality stations run by the New South Wales Department of Planning, Industry and Environment (DPIE) at Wollongong and Campbelltown West are also analysed, with the locations of all three observation sites shown in Figure 2.

Figure 2. Study area: Zoomed in map of MODIS firecount over Southeast Australia [48] (28° S– 40° S, 140° E– 152° E) from 2 December 2019 to 1 January 2020 where the yellow triangle shows the surface in situ and column CO (TCCON & NDACC) measurement site at Wollongong (UOW: 34.406° S, 150.879° E) and the NSW DPIE air quality station at Wollongong (34.417060° S, 150.887330° E), which is located close to the UOW site. The blue triangle shows NSW DPIE air quality station at Campbelltown West (34.066670° S, 150.795540° E).

2.2. In Situ FTIR

Ambient measurements of atmospheric trace gas mole fractions including CO at the Wollongong site are obtained from an in situ Fourier transform infrared (FTIR) trace gas analyser developed at the UOW, which is described in detail in Griffith et al. [49], Hammer et al. [50]. This analyser basically comprises of a FTIR spectrometer, which is a low-resolution Bruker IR cube, (1 cm^{-1}), combined with a White cell [51] and a thermoelectrically-cooled mercury cadmium telluride detector. The infrared source is modulated by passing through a calcium fluoride beamsplitter in a Michelson interferometer before entering the White cell, which reflects through an atmospheric sample in order to produce a folded path length of 24 m [49].

The in situ FTIR instrument measures wavenumbers from 1750 cm^{-1} to 5500 cm^{-1} and hence covers four broad spectral regions ($2097\text{--}2242\text{ cm}^{-1}$, $2150\text{--}2310\text{ cm}^{-1}$, $3001\text{--}3150\text{ cm}^{-1}$ and $3250\text{--}3775\text{ cm}^{-1}$) for retrieving the major trace gas mole fractions. CO is retrieved in the spectral region centred around 2143 cm^{-1} . The retrieval is performed using the non-linear least-squares program, Multiple Atmospheric Layer Transmission (MALT) [49,52]. Basically, MALT uses a model of the atmosphere and the instrument to produce a simulated transmittance spectrum using reference lines from the HITRAN database [53]. MALT then improves the spectral fit by iteratively changing parameters for the synthetic spectrum and comparing it to the measured spectrum until the residuals reach a minimum. MALT improves the simulated spectrum by altering the initial estimates of absorbing trace gas amounts, the instrument line shape and measured parameters such as temperature and pressure.

The analyser is calibrated periodically (typically monthly) against a suite of 3 or 4 reference gases provided by CSIRO/Gaslab (Melbourne, Australia) with assigned mole fractions on the scales used by the World Meteorological Organization-Global Atmosphere

Watch Programme (WMO-GAW) in situ network (WMO-GAW, 2020): WMO X2007 for CO₂, WMO X2004A for CH₄, WMO X2006A for N₂O, CSIRO94 for CO). Throughout most of the measurement record from Wollongong, the FTIR samples ambient air at a rate of 1 L min^{−1} during 23.5 h a day and the remaining 30 min is used for measuring a sample of constant composition from a gas cylinder (to check for any instrument drift). Trace gas mole fractions are retrieved from every spectrum, with a temporal resolution of 2 min (5 min earlier in the record). In order to assess any instrumental changes, a background spectrum under cell vacuum conditions is obtained periodically [54]. The instrument background remains stable except when there is any major instrument changes or movement occurs [54].

During the 2019–20 bushfire period, we use CO from the in situ FTIR located at the Wollongong measurement site measured at 3 min interval.

2.3. Total Carbon Column Observing Network (TCCON)

TCCON (<http://www.tcon.caltech.edu/>, accessed on 9 June 2021) is a global network of ground-based, high spectral resolution solar viewing Fourier transform spectrometers that record solar absorption spectra in the near-infrared. The principle is very similar to the in situ analyser, but with the sun providing the infrared source such that the pathlength for absorption is the whole atmosphere (or total column). TCCON aims to provide a continuous, accurate and precise measurement of trace gases, especially greenhouse gases, and other atmospheric constituents for a better understanding of the carbon cycle and to provide a primary validation dataset for satellite measurements [19].

The TCCON retrieval approach is based on the non-linear least-squares fitting algorithm known as “GFIT” [19], which scales an a priori profile to obtain the best spectral fit for the measured spectrum. The scaled profile is integrated to obtain total column abundance then converted to column-averaged dry air mole fraction (X_{gas}) by dividing by the column of dry air. In the TCCON retrieval approach, the total column amount of dry air is calculated as the ratio of the retrieved total column of O₂ to a known O₂ dry-air mole fraction of 0.2095. This strongly reduces the impact of surface pressure variations, avoids the need to account for H₂O in the column and thereby improves the precision of retrieved X_{gas} [19].

The TCCON consists of around 25 operational sites worldwide including Wollongong. The Wollongong TCCON site is co-located with the in situ FTIR at UOW (Figure 2). Under clear sky conditions, a total column measurement is performed approximately every 90 s. However, the measurement precision varies by site, which is generally <1% for X_{CO} [19]. Here we use X_{CO} and X_{CO_2} from the Wollongong TCCON site [55] as per the standard “GGG2014” TCCON data product [56].

2.4. Network for the Detection of Atmospheric Composition Change (NDACC)

NDACC (<http://www.ndacc.org>, accessed on 9 June 2021) is an international network of 70 stations around the world exclusively for providing high-quality measurements of atmospheric composition since 1991 [57]. The major objectives involved in NDACC network are providing a long term database of atmospheric composition and to better understand how changes and trends impact the atmosphere, establishing scientific links and feedback between atmospheric composition, climate change, and air quality, for filling the gaps made by satellite instruments and as a tool for validating other measurements especially, satellites. The ground-based FTIR instrument in Wollongong is dedicated to both TCCON & NDACC networks and has been continuously measuring trace gas concentrations for several years. The retrieval method for CO is as per the description provided in Zhou et al. [58].

In contrast to the TCCON network, the NDACC network uses mid-infrared spectra (MIR) from the FTIR measurements to retrieve the CO total columns. Wollongong NDACC data retrieval approach is based on the SFIT4 v9.4.4 retrieval algorithm (<https://wiki.ucar.edu/display/sfit4/Infrared+Working+Group+Retrieval+Code,+SFIT>, accessed on 9 June 2021), which produces the total and partial CO column based on three micro

windows in the 4.7 μm band [59]. The Wollongong data are retrieved using HITRAN 2008 line parameters [60], using apriori profiles from the mean of a 1980–2020 run of the atmospheric model “WACCM version 4” (and a 4 km Gaussian interlayer correlation) [61].

In this study, we use NDACC X_{CO} to extend/supplement the available TCCON X_{CO} data record at Wollongong. Zhou et al. [58] provides a detailed description of the comparison between TCCON and NDACC X_{CO} at six different sites across the world including Wollongong based on the study period: 2007–2017. According to their study, the absolute bias between the NDACC and the TCCON X_{CO} is within 2% at the Southern Hemispheric sites including Wollongong. However, we do not try to quantitatively use the NDACC and TCCON X_{CO} data interchangeably or as a combined dataset in this work in case there are remaining biases between the two datasets.

The NDACC total column CO was converted to the column-averaged dry-air mole fraction by the method detailed in Appendix A.

2.5. Air Quality Datasets

CO measurements (hourly averaged) from the New South Wales Department of Planning, Industry and Environment (DPIE) air quality stations in Wollongong and Campbelltown West were also used in this study (data available from: <https://www.dpie.nsw.gov.au/air-quality/air-quality-data-services>, accessed on 9 June 2021). At these stations CO is measured with a Teledyne API T300 using infrared spectroscopy, providing the ambient surface concentration by analysis of the sample air drawn into a cell. These monitoring stations record measurements continuously and provide air quality in hourly or less-frequent averages, and report data with a CO measurement precision of 0.1 ppm.

2.6. HYSPLIT Back Trajectories

In order to better understand the different air parcels in the surface and column, a detailed interpretation is performed using meteorology of air parcels by back trajectory analysis. Pre-calculated back trajectory analysis is performed using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model [62]. Short term NCEP Global Forecast System (GFS) forecast model output (<ftp://arlftp.arlhq.noaa.gov/pub/archives/gfs0p25>, accessed on 9 June 2021) at 0.25° horizontal resolution is used as the meteorological data for the trajectory analysis.

3. Results

Figure 3 presents the long term surface CO data record from the in situ FTIR measurements at Wollongong. The most prolonged enhancement of CO were measured during the 2019–20 bushfire period, which also corresponds to when the highest amounts are observed. While throughout the time series, regular peaks of around 2000 ppb are observed, during 2019–20 an episode of CO in excess of 3000 ppb was measured on 8 January 2020.

Figure 4 shows the long term data record of total column CO in Wollongong from 1996 to present from NDACC and from 2008 to present from TCCON. As for the in situ data, the 2019–20 bushfire signal in CO stands out as being the most extreme enhancement in the past 25 years of the CO data record, with the maximum X_{CO} reaching nearly 450 ppb. This clearly indicates the significance of the 2019–20 bushfires compared to the past events based on the available data record.

Figure 3. Time series of surface CO from April 2011–June 2020 recorded by the in situ FTIR at Wollongong.

Figure 4. Time series of long term column CO data record in Wollongong from NDACC (from May 1996) and TCCON (from July 2008–June 2020).

The seasonal cycle of multi-annual monthly median CO values in Figure 5 shows an approximate sinusoidal curve, which is driven by the CO sources and sinks. The influence of biomass burning is evident in the seasonal cycle, as the cause of the spring maximum (September–November) in the curve. The error bars shown in Figure 5 represent the inter annual variability of X_{CO} in Wollongong, which is relatively high at ~ 7.6 ppb, especially during the primary biomass burning season (September–November which corresponds to the biomass burning peak in Southern Hemisphere [13]), indicating the variability of biomass burning over the time period. Previous studies by e.g., [11,12,46] also indicated the impact of biomass burning in the seasonal cycle of CO at the Southern Hemispheric sites. The major loss mechanism of CO is through the reaction with photo-chemically produced hydroxyl radical.

Figure 5. Seasonal cycle of Wollongong column CO data calculated based on the monthly median with Median absolute deviation (MAD) of monthly data (as error bars) from the long term TCCON data record from July 2008 to June 2020.

The CO minimum occurs in late autumn (May), rather than summer when the OH production is at a maximum [63]. Several previous studies have reported that the chemical production of CO (oxidation of both CH₄ and NMVOCs) has a larger contribution in the Southern Hemisphere [64] and plays a major role in controlling the CO abundance in summer and autumn/early winter but has less influence in the primary biomass burning season [12].

It is well understood from previous studies [11] that long-range transport of biomass burning emissions plays a major role in variations of the geographic and seasonal distribution of CO in the southern hemisphere. Hence, the anomalous CO measurements in the time series (Figure 6) represent the effect of biomass burning and long-range transport in Wollongong. The CO seasonality in Wollongong was previously found to be highly influenced by biomass burning with no significant time lag between the surface and total column measurements [61].

Figure 6. Time series of (a) TCCON and NDACC X_{CO} anomalous values (Median + 3Median Absolute Deviation); (b) corresponding measurements of X_{CO_2} (TCCON) from July 2008 to June 2020 at Wollongong.

4. Discussion and Analysis of Anomalies

4.1. Wollongong X_{CO} and Surface CO Anomalies

To estimate the CO anomaly from the fires, the background CO needs to be quantified or estimated. Here, the background was calculated based on differing criteria for the surface concentrations and column-averaged dry-air mole fractions of carbon monoxide data. The background for the surface CO in Wollongong was determined based on the in situ FTIR data during the bushfire period (November 2019–February 2020). The background surface CO concentration at the site was calculated based on the 20th running percentile with a window length of 10 days (~75 ppb–130 ppb). Anomalies were subsequently determined based on the difference between the measurements and this background surface CO concentration. The lower bound of the percentile clearly represents the background surface CO concentration at the site, when the influence from sources other than fires is less pronounced. In addition, the 10-day window length period provides a robust measure of the background surface CO concentration as any pollution events from the city are likely to be of shorter time duration. The background of long-term column-averaged CO data (TCCON & NDACC) was performed based on the monthly medians from the long term data record since July 2008. Once defining the background, anomalous values are considered to be those more than 3 median average deviations (MAD) from the median.

Some of the most significant anomalous events illustrated in Figure 6 can be attributed to the major historical fires that occurred across Southern Australia. These include the “Black Saturday fires” in Victoria in 2009 [65] and the October 2013 fires in New South Wales [66]. However, in the X_{CO} anomalies, both of these events are dwarfed by the most recent 2019–20 bushfires. The CO enhancements observed during this bushfire episode were the largest ever recorded from Wollongong and highly variable over the time frame of hours and days. The corresponding enhancements in CO_2 (bottom panel of Figure 6) are less evident compared to CO because the CO_2 enhancements, while large in absolute magnitude, are small compared to the overall variability in the CO_2 timeseries.

Table 1 presents the comparison of X_{CO} metrics for 2019–20 bush fires with selected past Australian fire events based on the Fourier-Transform infrared Spectrometer (FTS) data record in Wollongong. In addition, the long-range transport from South American fires in October 2010 also results in marked CO anomalies in the column data record (mean X_{CO} anomaly = 27.25 ppb over 6 days). It is clearly evident that this fire season was highly anomalous in both intensity and duration compared to the past events in the long term data record. The duration of the Black Summer fire is ~6 times and the mean X_{CO} anomaly is around 2.5 times that of the past fire events indicated in Table 1.

Table 1. Comparison of X_{CO} metrics for 2019–20 fires with the past Australian fire events based on the FTS data record (hourly averaged).

Fire Event	Dates (YYYY/MM/DD)	Range (ppb)	Maximum (ppb)	Mean Anomaly (ppb)	Period (days)
Black Saturday	2009/02/05–2009/02/25	67	122	13	<20
NSW fires	2013/10/17–2013/10/26	147	201	12	~10
Black Summer	2019/11/15–2020/02/15	404	448	40	~90

4.2. How Well Do Enhancements in Total Column Amounts of CO at Wollongong Match the Enhancements in Surface CO?

We analyse here whether column measurements of CO can be used as a proxy for surface enhancements under fire-affected conditions, and therefore whether the column measurements can enable estimates of surface exposures to CO and other fire-related pollutants. We compare enhancements in the surface and column CO as determined based on the criteria already explained above. Figure 7 shows the hourly averaged surface in situ and column CO data in Wollongong from November 2019 to February 2020. Out of the 2904 possible hourly measurements over the 4-month period examined, 2640 h data are obtained from the in situ FTIR measurements after accounting for the smoothing performed for enhancements (for a window length of 10 days) and missing measurements, and coincident column and surface data are available ~20% of the time. We only address periods where both column and surface data are available and so there are only daytime data since the column measurements require direct sunlight. In addition, column measurements were also impacted during periods of heavy smoke, which blocks direct visible light, used to automate solar tracker. When enhancements are present, we can categorise the events into three possible scenarios:

- CO enhancements in the column only
- CO enhancements in both column & surface in situ
- CO enhancements in the surface in situ only

These are categorised based on the co-located Wollongong surface and column measurements only. The air quality sites are used to further investigate and understand the relationship between surface and column measurements, but due to their lower precision (100 ppb), these data are not the primary focus of the analysis.

The majority (approximately 71%) of the time that an enhancement is observed in the CO column over Wollongong, there is also an enhancement observed at the surface. As can be seen in Figure 7, the enhancements of surface in situ and column are often significantly different in their magnitude. Indeed, there is no robust correlation in the magnitudes of the column and surface enhancements. This will be investigated further in the next subsection. Approximately 15% of the time there is only enhancement in the column and not at the surface. That implies that none of the polluted air is reaching the surface, and suggests that the origin of the air masses in the surface in situ measurements and the column can be entirely different. The remaining 14% of the time enhancements are observed at the surface only, not in the column measurements. We will examine each of these event categories in more detail.

Figure 7. Time series of hourly averaged CO measurements at Wollongong from November 2019–February 2020: (a) surface in situ CO measurements at Wollongong air quality monitoring site (green squares), Campbelltown West air quality monitoring site (yellow triangles) and Wollongong in situ FTIR (red crosses); (b) column CO measurements (TCCON and NDACC) (black dots). The colour coded vertical bars show the different categories of events: CO enhancements in the column only (purple), CO enhancements in the surface in situ only (blue) and CO enhancements in both column & surface in situ (grey).

4.2.1. CO Enhancements in the Column Only

During the Black Summer fires, 15% of the time that enhanced CO was measured this was observed in the column only with no corresponding enhancement evident at the surface. For example, for the event on December 1 2019, as shown in Figure 8, the hourly column CO values (~80 ppb–160 ppb) are high compared to the corresponding surface CO measurements. The Wollongong surface in situ measurements show no enhancements above a relatively constant background (~100 ppb). The Wollongong air quality measurements are also low, varying between 100 and 200 ppb throughout the day. In contrast, the Campbelltown West air quality site (elevation = 112 m) follows a similar enhancement pattern to the column measurements.

Figure 8 also presents the HYSPLIT back trajectories for 1 December 2019 at different altitudes to approximately represent the air mass histories for the column (3000 m) and surface (30 m). The 24 h back trajectories at the representative surface altitude show a mix of low level winds dominated by southeasterly winds from the sea in Wollongong, and westerly winds from inland. These westerly winds bring airmasses from the burned regions later during the day and explains the highest surface enhancements during the event. From the MODIS visible image of smoke, along with the fire locations, the significant burning surrounding Wollongong is evident, stretching from north to south of the site. Therefore, the higher altitude trajectories, dominated by the westerlies that travel over the burned regions, are responsible for the higher column enhancements during the event.

Similar events under this category also occurred on 9 December 2019, 28 December 2019 (see Appendix B, Figure A1) and 10 January 2020. All these events follow a similar pattern with significant CO enhancements in the column and comparatively low enhancements at the surface. In order to better understand the reason for the occurrence of these events, we look at the effect of topography and local meteorology on the site. Wollongong is located on a narrow coastal strip sandwiched between the sea and the escarpment. These complex topographical features at the site suggest the possibility of the formation of stacked layers, with a well-mixed layer above the escarpment to the west of Wollongong, and comparatively cleaner air over Wollongong itself due to the proximity of the sea [67].

Figure 8. 1 December 2019: 24 h NOAA HYSPPLIT back trajectories (UTC) at (a) 30 m and (b) 3000 m; (c) Hourly averaged surface in situ CO measurements at Wollongong air quality monitoring site (green squares), Campbelltown West air quality monitoring site (yellow triangles) and Wollongong in situ FTIR (red crosses). Black and grey dots show hourly averaged column CO measurements (TCCON & NDACC) and all native temporal resolution column measurements, respectively (in AEST: UTC + 10 h). (d) MODIS hotspot image with thermal anomalies [68] (zoomed in for southeast Australia). The UOW measurement site is marked with a yellow triangle.

In order to better understand this, we look at the surface CO observed over the Campbelltown West air quality station, situated west of the escarpment and north-west of Wollongong, ~55.5 km from Wollongong. For this site, we observe occasions where high column CO values occur in conjunction with relatively high surface CO observed over Campbelltown (situated well above the site in Wollongong), but when the Wollongong surface CO shows only small enhancements. Figure 9 shows the surface and column CO enhancements on 14 December 2019. This event is marked by relatively higher surface CO enhancements in Campbelltown compared to the other measurements. Interestingly, we can see that the surface enhancements associated with Campbelltown strongly correlate with the corresponding enhancements in the column. Surface trajectories for Wollongong show air masses passing over the clean ocean, in contrast to the column trajectories that originate from inland. This highlights the role of topography and local meteorological effects in determining the representative surface enhancements at the site.

Figure 9. 14 December 2019: 24 h NOAA HYSPLIT back trajectories (UTC) at (a) 30 m and (b) 3000 m; (c) Hourly averaged surface in situ CO measurements at Wollongong air quality monitoring site (green squares), Campbelltown West air quality monitoring site (yellow triangles) and Wollongong in situ FTIR (red crosses). Black and grey dots show hourly averaged column CO measurements (TCCON & NDACC) and all native temporal resolution column measurements, respectively (in AEST: UTC + 10 h). (d) MODIS hotspot image with thermal anomalies [68] (zoomed in for southeast Australia). The UOW measurement site is marked with a yellow triangle.

4.2.2. CO Enhancements in Both the Column & Surface In Situ

This is the most prevalent category of events that occurred during the bushfire period, where there is an occurrence of simultaneous enhancements in both the column and the surface. From the available data record, the events on 19 and 21 December and 1 January can be classified under this category. A recent study by Khaykin et al. [69] has shown that the 2019–20 wildfires have caused exceptionally strong Pyro-cumulonimbus (pyroCb) activity in the southeast of the continent, which is proportional to the strongest volcanic eruption in the last 25 years, with the strongest PyroCb outbreak occurring on 31 December/1 January. This corresponds with the highest column averaged CO enhancements observed at Wollongong of more than 250 ppb.

Figure 10 presents the surface and column CO enhancements at the study area during the event on 31 December 2019 to 1 January 2020. Large enhancements can clearly be seen in both the surface and column values. The surface in situ measurements at Wollongong show a peak of ~1000 ppb around midday. This clearly implies the intensity of the fires and the smoke traversed and mixed near the site as seen in the MODIS smoke image, where the most intense visible plume is over Wollongong and the ocean to the east. The back trajectories show that the site is influenced by a mix of low-level winds from the southeast and northeast predominantly from inland rather than from the sea. In contrast to the prevailing influence of cleaner air surrounding the site, the ambient air at the site is well mixed with the smoke during this event. The column trajectories presented in Figure 10 show the smoke enriched air parcels coincide with the fire locations in the MODIS smoke image reaching from the west.

Figure 10. 1 January 2020: 24 h NOAA HYSPLIT back trajectories (UTC) at (a) 30 m and (b) 3000 m; (c) Hourly averaged surface in situ CO measurements at Wollongong air quality monitoring site (green squares), Campbelltown West air quality monitoring site (yellow triangles) and Wollongong in situ FTIR (red crosses). Black and grey dots show hourly averaged column CO measurements (TCCON & NDACC) and all native temporal resolution column measurements, respectively (in AEST: UTC + 10 h). (d) MODIS hotspot image with thermal anomalies [68] (zoomed in for southeast Australia). The UOW measurement site is marked with a yellow triangle.

The category of events where there are enhancements in both the column and surface measurements of CO is the most commonly observed during the bushfire period but the magnitude of enhancements is usually different in the column and in the surface. A correlation analysis shows that there is little correlation between the surface and column, $r^2 = 0.25$ (See Appendix C—Figure A2). Column measurements therefore cannot be considered to be representative of surface abundances of CO and other associated fire products under these conditions.

4.2.3. CO Enhancements in the Surface In Situ Only

The third event category during the wildfires is when there are large enhancements of CO at the surface and relatively low enhancements in the column. The best example of this is the event on 5 December 2019, shown in Figure 11. While in this case, no enhancements are seen in either the column or surface CO measurements at Wollongong, the Campbelltown West site records high enhancements throughout the period that column measurements are available. The Campbelltown West site is only 55 km from Wollongong and therefore within the same pixel for many satellites such as MOPITT. Later in the day, there is better agreement between all the surface CO measurements with considerable enhancements measured. The representative surface trajectories shown in Figure 11a clearly indicate the slow moving trajectories aligned with the burned regions in the MODIS smoke image, which explain the nighttime buildup of the surface CO. At this time, burning and the consequent observed visible smoke plumes were not as widespread and more localised than in some of the later events around the end of the year. In contrast, the column trajectories are highly influenced by the air parcels that are comparatively “cleaner”, from further south in the continent.

Figure 11. 5 December 2019: 24 h NOAA HYSPLIT back trajectories (UTC) at (a) 30 m and (b) 3000 m; (c) Hourly averaged surface in situ CO measurements at Wollongong air quality monitoring site (green squares), Campbelltown West air quality monitoring site (yellow triangles) and Wollongong in situ FTIR (red crosses). Black and grey dots show hourly averaged column CO measurements (TCCON & NDACC) and all native temporal resolution column measurements, respectively (in AEST: UTC + 10 h). (d) MODIS hotspot image with thermal anomalies [68] (zoomed in for southeast Australia). The UOW measurement site is marked with a yellow triangle.

A limitation of this study is a lack of column data during some of the fire events that impacted the site. In this category, of surface enhancements only, we have identified a few events where there are exceptionally large enhancements in the surface, with CO reaching mole fractions of ~3–4 ppm. However, column data are not available for these particular events due to smoke blocking the visible light used to automate the solar tracker. Under these conditions, the automated collection software fails to operate and so ground-based column measurements of CO were not taken.

Figure 12 shows the surface enhancements on 8 January from the available datasets. The data recorded by the in situ FTIR and at the Wollongong air quality site show relatively high CO enhancements from the early morning hours, whereas the Campbelltown West site does not observe the same magnitude of enhancement until later in the morning. Interestingly, the Wollongong surface datasets show a similar pattern of surface CO enhancements with a significant peak in magnitude of around 3.5 ppm during middle of the day. While the timing of the peak is similar, we can see that the magnitude of surface enhancements in the Campbelltown West data is significantly lower than the measurements in Wollongong during the event. The HYSPLIT trajectories indicate that the surface air masses are originating from south, along the coast where significant biomass burning plumes can be seen in the MODIS visible image. In contrast, the representative column trajectories show that the air masses are predominantly from the west and north-west. While there is less visible smoke in this sector, the MODIS image indicates that from the Wollongong site clear skies would not be present during this period, and hence column measurements are not possible. In addition, there was a PyroCb outbreak with stratospheric impact that happened on the previous day (7 January), with a horizontal extent of 6.1 million km² [69], which might have caused a major impact on the atmospheric column measurements. A similar event was recorded on 10 December.

Figure 12. 8 January 2020: 24 h NOAA HYSPLIT back trajectories (UTC) at (a) 30 m and (b) 3000 m; (c) Hourly averaged surface in situ CO measurements at Wollongong air quality monitoring site (green squares), Campbelltown West air quality monitoring site (yellow triangles) and Wollongong in situ FTIR (red crosses). Black and grey dots show hourly averaged column CO measurements (TCCON & NDACC) and all native temporal resolution column measurements, respectively (in AEST: UTC + 10 h). (d) MODIS hotspot image with thermal anomalies [68] (zoomed in for southeast Australia). The UOW measurement site is marked with a yellow triangle.

5. Summary and Conclusions

The 2019–20 bushfires were unprecedented in intensity, duration and area burned in Southeast Australia. This anomalous duration and intensity is borne out by the FTS data record in Wollongong, which saw the highest recorded column CO during the time series, and an unprecedented length of enhancement in both column and surface measurements due to burning. We found that when both column and surface data are available, 70% of enhancement events saw enhancements in both the surface in situ and column CO at Wollongong, but the correlation in the magnitudes of these enhancements is weak. This implies that we cannot robustly infer surface in situ CO and subsequent exposure based on ground-based or satellite-based column measurements. The topography at Wollongong, combined with atmospheric dynamics, potentially exacerbates differences in the surface and column measurements. We conclude that satellite column amounts are unlikely to provide an accurate reflection of exposure at the ground during major fires like the 2019–2020 bushfires, especially in a narrow coastal city like Wollongong, where the land is sandwiched between the sea and the escarpment. This indicates the complex topographical features near Wollongong combined with the meteorology (as indicated by the back trajectories), result in differences in surface and column enhancements. Since most of the Australian population lives in coastal regions, like Sydney and Melbourne, which also sit in basins with similar, albeit less pronounced, topological features, it is unlikely the column CO measurements can be used to infer population exposure for large amounts of the Australian population.

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Data Availability Statement: TCCON data at Wollongong, Australia are available publicly through the TCCON data archive, <https://tccodata.org/>, accessed on 9 June 2021. NDACC data at Wollongong is available from the NDACC website (<http://www.ndacc.org>, accessed on 9 June 2021). The in situ FTIR data at Wollongong until 2015 are available online and the rest of the data is under preparation, <https://doi.org/10.1594/PANGAEA.848263>, accessed on 9 June 2021. Air quality measurements of CO at Campbelltown West & Wollongong are publicly available, <https://www.dpie.nsw.gov.au/air-quality/air-quality-data-services/data-download-facility>, accessed on 9 June 2021.

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Appendix A. Conversion of CO Total Column to X_{CO}

In order, to make the column measurements consistent with the TCCON reported measurements, the NDACC total column CO was converted to the column-averaged dry-air mole fraction. Equation (A1) below was used to convert the retrieved total CO column to the X_{gas} .

$$X_{i,dry} = \frac{VC_i}{\frac{P_s}{m_{dryair} \cdot g(p)} - VC_{H_2O} \frac{m_{H_2O}}{m_{dryair}}}, \quad (A1)$$

where:

$X_{i,dry}$ = Column averaged dry air mole fraction;

VC_i = Vertical column of gas i ;

P_s = Surface Pressure (mb);

$g(p)$ = Absorber weighted gravitational acceleration;

m_{dryair} = Molecular mass of dry air, $28.964 \text{ g mol}^{-1}$; and

m_{H_2O} = Molecular mass of H_2O , 18.02 g mol^{-1} .

Appendix B. CO Enhancements in the Column with Minor Enhancements at the Surface

Several events occur where the column enhancements are relatively large compared to those measured at the surface. For example, on 28 December, as shown in Figure A1, the column values range from 127 ppb to 200 ppb. On the same day, the in situ FTIR shows a small double-peak pattern of enhancements first, in the middle of the day reaching up to 200 ppb, and second, during the end of the day around 180 ppb. Also, the Campbelltown CO data has shown similar enhancements to around 400 ppb before and after the first peak enhancements recorded by the in situ FTIR, and depicts an agreement with the corresponding column enhancements that happened during the day.

The 24 h back trajectories at the surface altitude clearly indicate the dominance of northeasterly winds from the sea in Wollongong. Wollongong is dominated by northeasterly winds especially during spring and summer, associated with the peak in sea breeze influence [54]. The surface back trajectory also indicates that the air parcels reaching the surface are highly influenced by the sea during the beginning of the day and later are slightly mixed with air parcels from the burned regions, explaining the small double peak enhancements recorded by the in situ FTIR. Hence, the column trajectories indicate that the air parcels are contaminated by the presence of smoke and exhibiting a slow anticyclonic circulation (which normally helps in recirculating and trapping local and transported pollution especially from Sydney during spring/summer) [46], which contributed to the higher column enhancements observed during the event.

Figure A1. As for Figure 8, but for 28 December 2019.

Appendix C. Additional Plots

Figure A2. Correlation plot of total column CO enhancement vs surface enhancement.

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