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When smoke comes to town: The impact of biomass burning smoke on air quality

Melita Keywood [a, *], Martin Cope [a], C.P. Mick Meyer [a], Yoshi Iinuma [b], Kathryn Emmerson [a]

[a] CSIRO Oceans and Atmosphere Flagship, PMB1 Aspendale, Victoria 3195 Australia
[b] Leibniz-Institut fuer Troposphaerenforschung (TROPOS), Permoserstr. 15, 04318 Leipzig, Germany

HIGHLIGHTS

- Plumes that impacted Aspendale resulted in elevated concentrations of particles and gases.
- Old plumes showed evidence of more photochemistry than young plumes.
- Speciated SOA compounds made up a greater fraction of speciated organic mass old plumes.
- Speciated biomass burning compounds dominated the speciated organic mass in the young plumes.
- Smoke plumes resulted in elevated concentrations of particles and gases.

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ABSTRACT

Biomass burning aerosols influence the radiative balance of the earth-atmosphere system. They also reduce visibility and impact human health. In addition, trace gases and aerosols emitted to the atmosphere during large biomass burning episodes may have a significant effect on atmospheric chemistry due to the presence of reactive species.

Six hundred and ninety wildfires burned more than one million hectares in Victoria, Australia between December 2006 and February 2007. Thick smoke haze was transported to Melbourne (population 3.9 million) on several occasions, causing PM_{10} (particulate mass less than 10 μm in diameter) concentrations to exceed 200 μg m^{-3}. The presence of elevated total secondary organic aerosol (SOA) and speciated SOA compounds (including pinene and cineole oxidation products), O_3, and the larger aerosol mode diameter during smoke impacted periods indicated the presence of photochemical oxidation within the plume. The presence of organosulfate compounds and nitro-oxy organosulfate compounds indicated oxidation may have occurred in the presence of acidic seed aerosol and that oxidation may also have occurred at night.

Older smoke plumes (aged 30 h) displayed higher concentrations of a number of gaseous and aerosol species relative to the younger smoke plumes (aged 3 h). SOA compounds made up a greater fraction of speciated organic mass in the old plume than in the young plume where speciated biomass burning compounds dominated. Cineole oxidation products made up a greater fraction of the speciated SOA compounds in the old plume while pinene oxidation products made up a greater fraction of the total SOA speciated mass in the samples from the young plume. This may be a result of the slower reaction rate of cineole with OH. Organosulfate compounds and nitro-oxy organosulfate compounds made up greater fractions of the speciated SOA mass in the old plume consistent with the production of nitro-oxy organosulfate compounds under night time conditions in the presence of acidic seed.

These results suggest that enhanced photochemical activity occurs in smoke plumes and can significantly change the composition and microphysical properties of aerosol, potentially leading to changes in the optical and thus radiative properties of the aerosol.

* Corresponding author.
E-mail address: melita.keywood@csiro.au (M. Keywood).

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1. Introduction

Ito and Penner (2005), estimate that the global amount of biomass burned in the year 2000 was 5.6 Pg, releasing 38.3 Tg of $PM_{2.5}$ (particulate matter with an aerodynamic diameter of 2.5 μm or less). Biomass burning aerosols influence the radiative balance of the earth-atmosphere system directly through the scattering and absorption of radiation, and indirectly through their influence on cloud microphysical processes, and therefore constitute an important forcing on the climate (e.g., Rotstayn and Penner, 2001). They also reduce visibility, influence atmospheric photochemistry and can be inhaled into the deepest parts of the lungs, so that they can have a significant effect on human health (Pope and Dockery, 2006).

Biomass burning emissions have a significant effect on atmospheric chemistry due to the presence of reactive species. In fresh fire plumes large fluxes of emitted oxides of nitrogen (NO_x) titrate ozone (O_3) resulting in low O_3 concentrations (Crutzen and Andreae, 1990; Monks et al., 2009; Amiridis et al., 2012). Aged plumes have high concentrations of secondary pollutants such as O_3, and a higher proportion of oxygenated species including secondary aerosols (sulfates, nitrates and organics) (Yan et al., 2008). For example Lee et al. (2008) identified elevated $PM_{2.5}$ and O_3 in Atlanta associated with smoke from a prescribed-burning fire on the outskirts of the city. Ratios of organic carbon (OC)/elemental carbon (EC) and OC/potassium suggested the OC content of $PM_{2.5}$ had a significant fraction of secondary components including water soluble hydrophobic compounds, possibly derived from oxidation of isoprenoid emissions that have been shown to be enhanced under high temperatures associated with forest fires (Alessio et al., 2004).

Australia experiences wildfires on an annual basis. In most years fires are restricted to the tropical savanna forests of northern Australia, contributing about 8% of global carbon emissions from biomass burning (Kasischke and Penner, 2004), with 90–95% of this resulting from wildfires. However, there have been many wildfires in southern Australia's history including the Black Saturday fires of February 2009, that burned 450,000 hectares and claimed 179 lives (Cruz et al., 2012).

The association between health outcomes and forest fire smoke is complex, and includes worsening of lung function and respiratory diseases (Naeher et al., 2007; Morgan et al., 2010; Martin et al., 2013), increased death rates in affected populations (Johnston et al., 2011), and increased incidence of heart disease (Dennekamp et al., 2015). The measured concentration of particles can be a marker for the complex mixture of different constituents (gas and particles) that make up smoke (Naeher et al., 2007). Studies have shown that PM from forest fires produce greater lung toxicity in laboratory animals than the equivalent mass of particulate matter from urban sources (Wegesser et al., 2009).

The summer of 2006/2007 (December 2006–February 2007) was marked by the longest recorded period of fires in Victoria's history. Approximately 690 separate wildfires, including the major Great Divide Fire (GDF) burned more than one million hectares over 69 days. Thick smoke haze was transported to the Melbourne central business district (CBD) on several occasions (Fig. 1) and PM_{10} concentrations at several air quality monitoring stations peaked at over 200 μg m^{-3} (four times the National Environment Protection Measure PM_{10} 24 h standard).

During this period, a comprehensive suite of air quality measurements was carried out at Aspendale, located 25 km south of the Melbourne CBD including detailed aerosol microphysical and chemical composition measurements.

The data collected provide a unique opportunity to investigate the effect of the smoke on air quality in the Melbourne airshed and the changes in aerosol microphysical and chemical properties that occurred as the smoke plume was transported to Melbourne and interacted with the urban plume. The interaction between smoke and urban plumes has important public health implications, because of the injection of primary particles and gases into the airshed, and the secondary products formed as the plume ages. Although Keywood et al. (2011) have estimated the contribution of secondary organic aerosol (SOA) to the Melbourne airshed, the organic composition of aerosol in Australian cities is not well characterised.

Here we examine the chemical and physical properties of the smoke plume as it impacted the Melbourne air shed and discuss its impact on air quality over the city. We estimate total constituent emissions from the fires and determine the age of the air masses using a chemical transport model. Finally we investigate the changes in the aerosol composition and microphysical properties associated with the photochemical aging of the smoke plumes.

2. Materials and methods

2.1. Air quality and meteorology measurements

Measurements were carried out at Aspendale which is located 25 km south of the Melbourne CBD. Measurements included aerosol chemical composition and microphysical properties, concentrations of a number of gaseous species and meteorological parameters.

Table 1 lists the measurement frequency and methodologies used in this study, and the time series of a number of aerosol and gaseous species are shown in Figure S3 of the Supplementary material. Further details of the instrumentation used can also be found in the Supplementary material.

The methodology used to estimate the SOA concentrations is also presented in the Supplementary material as are the details of methodologies used to measure a series of biomass burning and biogenic SOA marker compounds in the PM_{10} samples.

2.2. Chemical transport modelling

$PM_{2.5}$ emissions from the fires burning in Victoria between December 2006 and February 2007 were determined using a variant of the methodology presented in Meyer et al. (2008), and described in detail in the Supplementary material.

Table 2 shows that the total emissions from the GDF for December 2006 are estimated to exceed those from the anthropogenic sources by more than a factor of 100 for NO_x, carbon monoxide and $PM_{2.5}$.

The transport and chemical transformation of emissions from the Victorian region (both anthropogenic and fire-related) were modelled using the TAPM–Chemical Transport Model (TAPM-CTM) (Cope et al., 2004). A detailed description of the system is presented in the Supplementary material. Fig. 1 shows an example of the modelled plume compared with a MODIS image of the same plume.

2.3. Plume age determination

The modelling system was used to estimate plume ages of air parcels passing over Aspendale between 2 and 31 December 2006. Details of the methodology used to determine plume age are presented in the Supplementary material.

Fig. 2 shows the time series of the calculated plume age for the GDF simulation. Because we are treating NO as the tracer the figure shows the mean plume age of the combined Melbourne and GDF plumes.

Other studies have used satellite imagery to estimate plume