



Ozone and particulate matter enhancements from regional wildfires observed at Mount Bachelor during 2004–2011



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HIGHLIGHTS

- We report enhancement ratios for 32 wildfires in the western US and Canada.
- We find significant secondary organic aerosol formation in plumes aged <2 days.
- Ozone production may be impacted by the altitude of plume transport.
- The presence of urban emissions enhances ozone production in wildfire plumes.

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ABSTRACT

We report observations of normalized enhancement ratios (NER) for 32 wildfires measured at Mount Bachelor Observatory in central Oregon during June–September 2004–2011. All 32 plumes resulted from wildfires originating in the western United States and Canada. The observed NER of PM₁ (particulate matter < 1 micron) to carbon monoxide ($\Delta\text{PM}_1/\Delta\text{CO}$) ranged from 0.06 to 0.42 $\mu\text{g m}^{-3}$ ppbv⁻¹. The NER of ozone to CO ($\Delta\text{O}_3/\Delta\text{CO}$) ranged from 0.01 to 0.51 ppbv ppbv⁻¹ for the 13 observed plumes with a significant $\Delta\text{O}_3/\Delta\text{CO}$ NER ($p \leq 0.01$, $R^2 \geq 0.30$). For wildfire plumes transported <540 km, or approximately <2 days, the $\Delta\text{PM}_1/\Delta\text{CO}$ NER is found to increase with increasing distance, suggesting that there is significant secondary organic aerosol (SOA) production in these plumes. However, two plumes transported over greater time periods have relatively low $\Delta\text{PM}_1/\Delta\text{CO}$ NER, indicating that PM₁ loss is greater than SOA production in these plumes. Of the three plumes transported the longest distance to MBO, only two have significant O₃ production. These two plumes were transported in boundary layer air masses, while the third was transported in a free tropospheric air mass, suggesting that conversion of nitrogen oxides (NO_x) to peroxyacetyl nitrate (PAN) may be a factor affecting O₃ production in these plumes. Two wildfire plumes are mixed with urban emissions from the Seattle/Tacoma metropolitan area, and have relatively higher $\Delta\text{O}_3/\Delta\text{CO}$ NER than other wildfire plumes transported over similar distances.

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Abbreviations: BC, British Columbia; BL, boundary layer; CA, California; CO, carbon monoxide; EDAS, Eta Data Assimilation System; FIRMS, Fire Information for Resource Management System; FT, free troposphere; GDAS, Global Data Assimilation System; H₂O(g), water vapor; HYSPLIT, Hybrid Single Particle Lagrangian Integrated Trajectory; MBO, Mount Bachelor Observatory; MODIS, Moderate Resolution Imaging Spectroradiometer; MDL, method detection limit; NAAPS, Navy Aerosol Analysis and Prediction System; NER, normalized enhancement ratio; NO_x, nitrogen oxides; OA, organic aerosol; OR, Oregon; O₃, ozone; PAN, peroxyacetyl nitrate; PM₁, particulate matter < 1 micron; PM_{2.5}, particulate matter < 2.5 microns; PNW, Pacific Northwest; SOA, secondary organic aerosol; STP, standard temperature and pressure.

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1. Introduction

Wildfires are a global source of numerous primary and secondary air pollutants (Akagi et al., 2011; Andreae and Merlet, 2001). Many factors impact the relative proportions of trace gases and aerosols emitted from wildfires and produced during plume transport including fire size, fuel type and loading, fire combustion efficiency and meteorological conditions (Akagi et al., 2011; Jaffe and Wigder, 2012). A number of recent aircraft and in situ studies report fire emissions and downwind mixing ratios of ozone (O₃) and PM_{2.5} (particulate matter < 2.5 microns in diameter) in the western United States (US) and Canada (Akagi et al., 2012; Burling et al., 2011; Hecobian et al., 2011; Jaffe et al., 2008; McKendry et al., 2011; Singh et al., 2012). These results demonstrate large variability

in the emissions and subsequent photochemistry of temperate and boreal fire plumes.

Production of secondary air pollutants, especially O₃ and secondary organic aerosol (SOA), are important to understanding the relative concentrations of these pollutants for the global budget and regional air quality assessments (Akagi et al., 2012). Ozone is created from the interaction of nitrogen oxides (NO_x) and non-methane organic compounds in the presence of sunlight. Wildfire O₃ production is highly variable, although O₃ enhancements in wildfire plumes (relative to carbon monoxide (CO) enhancements) generally increase with increasing plume age. In a recent literature review, Jaffe and Wigder (2012) found that the enhancement ratio of O₃ relative to CO ($\Delta O_3/\Delta CO$) in boreal and temperate wildfires was on average 0.018, 0.15 and 0.22 ppbv ppbv⁻¹ for plumes aged 1–2 days, 2–5 days and ≥ 5 days, respectively. Additionally, Real et al. (2007) found that high concentrations of aerosols in wildfire plumes can either decrease or increase net O₃ mixing ratios, as the presence of aerosols affects the rates of both photochemical O₃ production and loss.

The limiting factor in wildfire O₃ production is typically NO_x (Jaffe and Wigder, 2012; Mauzerall et al., 1998; Singh et al., 2012). Mixing ratios of NO_x in fire plumes are impacted by the fire combustion efficiency, fuel nitrogen content, and photochemical production and loss of species such as peroxyacetyl nitrate (PAN). Conversion of NO_x to PAN has been shown to account for a large fraction of rapid NO_x loss in boreal (Alvarado et al., 2010) and temperate (Akagi et al., 2012) fire plumes. Wildfire plumes that are mixed with urban emissions have higher O₃ mixing ratios, partly due to the addition of urban NO_x emissions (Akagi et al., 2013; Singh et al., 2012). Recent studies have also shown that NO_x emissions from wildfires may be enhanced due to deposition of urban emissions prior to the fire ignition (Burling et al., 2011; Yokelson et al., 2011).

Particulate matter (PM) is both a primary and secondary pollutant from wildfires. Approximately 80–90% of particulate aerosol volume in biomass burning emissions is in the accumulation mode (PM₁: PM < 1 micron in diameter; Reid et al., 2005), although many wildfire studies report PM_{2.5} concentrations, which is a measurement used in US and Canadian regulations. A laboratory study of fire-integrated biomass burning emissions showed that PM_{2.5} emission factors depend on both the type of fuel burned and the fire combustion efficiency (McMeeking et al., 2009). The rate of secondary organic aerosol (SOA) production is a major factor affecting the evolution of PM concentrations in wildfire plumes (Akagi et al., 2012). In a recent study of a prescribed chaparral fire in California, Akagi et al. (2012) found that the enhancement of PM_{2.5} (relative to the enhancement of carbon dioxide) did not increase during the first 4–4.5 h of plume aging. Differently, a study of a plume in Mexico as part of the MILAGRO campaign showed that PM_{2.5} concentrations increased by a factor of 2.6 after approximately 1.4 h (Yokelson et al., 2009). The authors also found that the enhancement ratio of black carbon to CO was conserved as the plume aged, but the enhancement ratio of organic aerosols (OA) to CO strongly increased, signaling that the increase in PM_{2.5} is attributable to SOA production. Additionally, in a study of OA data from several aircraft campaigns, Jolleys et al. (2012) found that there was little SOA production in aged biomass burning plumes, although the authors noted a wide variation in measured OA enhancements among studies.

In this study, we analyze the enhancements of PM₁ and O₃ in 32 wildfire plumes observed at Mount Bachelor Observatory (MBO) during June–September 2004–2011. All 32 plumes result from fires in the western US and Canada, allowing us to study the variations in PM₁ and O₃ enhancements from regional wildfires. This study presents the largest dataset of wildfire plumes in the US Pacific

Northwest (PNW) to date and is the first comprehensive analysis of wildfire plumes observed at MBO.

2. Methods

2.1. Mount Bachelor Observatory site description and measurements

MBO is a mountaintop site located in central Oregon, US (43.98° N, 121.69° W, 2763 m above sea level) and is well positioned to observe long-range transport of Asian industrial and biomass burning plumes, as well as North American wildfires (Ambrose et al., 2011; Finley et al., 2009; Jaffe et al., 2005; Weiss-Penzias et al., 2007, 2006). During 2004–2011, measurements of CO, sub-micron aerosol scattering, O₃ and meteorological parameters were taken at MBO during the fire season (June–September). Carbon monoxide was sampled with a Thermo Electron Corporation non-dispersive infrared analyzer and O₃ with a Dasibi UV absorbance instrument (Weiss-Penzias et al., 2006). During 2004–2009 sub-micron dry aerosol scattering (at 535 nm) was measured with a Radiance Research M903 nephelometer (Weiss-Penzias et al., 2006). Submicron dry aerosol scattering (at 450, 550 and 700 nm) was measured with a TSI Model 3563 nephelometer during 2010–2011 (Fischer et al., 2010), and the 550 nm scattering coefficients are used in this analysis. For consistency, the 550 nm aerosol scattering coefficients were adjusted to 535 nm following the method of Müller et al. (2011). Briefly, this method uses the equation $\sigma_{535} = \sigma_{550} * (\lambda_{550}/\lambda_{535})^{2.450,550}$, where σ refers to a scattering coefficient, λ refers to a wavelength and α is the Angstrom Exponent calculated at the two wavelengths specified. Aerosol scattering data was corrected to STP ($T = 273.15$ K, $P = 1013.25$ kPa) and then converted to mass concentrations ($\mu\text{g m}^{-3}$) using a dry aerosol scattering efficiency of $3.6 \text{ m}^2 \text{ g}^{-1}$ (Hand and Malm, 2007). This dry aerosol scattering efficiency was used because it was determined to be the best for mixed composition aerosols by the Hand and Malm (2007) review article; however, it should be noted that mass scattering efficiencies have been shown to change as a plume ages (Akagi et al., 2012) and that there are uncertainties in all mass scattering efficiencies. Water vapor (H₂O(g)) mixing ratios were calculated from measurements of temperature and relative humidity taken with a Campbell Scientific HM45C. All trace gas, aerosol and meteorological observations analyzed were one-hour averages, and concentrations below the method detection limits (MDL) were set to one-half of the MDL.

2.2. Wildfire identification

Wildfires were identified in the MBO dataset using a combination of PM₁ and CO observations, satellite data and trajectory models. First, potential fire time periods were identified based on three criteria: (1) PM₁ and CO concentrations were elevated above the background, (2) for at least one hour, ambient aerosol scattering $\geq 20 \text{ Mm}^{-1}$ and CO ≥ 125 ppbv, and (3) there was a strong correlation ($R^2 \geq 0.70$) between the aerosol scattering and CO observations. Next, Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) model backward air mass trajectories were initiated from MBO at intervals throughout each of the potential fire periods (Draxler, 1999; Draxler and Hess, 1997, 1998). Both 10-day backward trajectories using the 1° resolution Global Data Assimilation System (GDAS) meteorological data and 3-day backward trajectories using the 40 km resolution US Eta Data Assimilation System (EDAS) meteorological data were calculated. Finally, several satellite products and models were used to identify fires or fire plumes intersecting the calculated trajectory paths. These products were:

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