



Wildland fire emissions, carbon, and climate: Plume rise, atmospheric transport, and chemistry processes



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ABSTRACT

This paper provides an overview and summary of the current state of knowledge regarding critical atmospheric processes that affect the distribution and concentrations of greenhouse gases and aerosols emitted from wildland fires or produced through subsequent chemical reactions in the atmosphere. These critical atmospheric processes include the dynamics of plume rise, chemical reactions involving smoke plume constituents, the long-range transport of smoke plumes, and the potential transport of gases and aerosols from wildland fires into the stratosphere. In the area of plume-rise dynamics, synthesis information is provided on (1) the relevance of plume height for assessing impacts of gases and aerosol from wildland fires on the climate system, (2) recent scientific advances in understanding the role of multiple updraft cores in plume behavior, and (3) some of the current modeling tools and remote sensing monitoring techniques available for predicting and measuring smoke plume heights. In the area of atmospheric chemistry associated with wildland fire emissions, synthesis information is provided on what is currently known about the atmospheric fate of wildland fire smoke-plume constituents and the relationship of their atmospheric chemistry to radiative forcing. Synthesis information related to long-range atmospheric transport of wildland fire emissions is presented and summarizes many of the recent published observational and modeling studies that provide clear evidence of intercontinental, continental, and regional transport of North American fire emissions, including black carbon, to locations far-removed from the fire-event locations. Recent studies are also highlighted that examined the significance of troposphere-stratosphere exchange processes, which can result in the transport of greenhouse gases and aerosols from North American wildland fires into the stratosphere where they can remain for very long periods of time and alter the radiative balance and typical chemical reactions that occur there. Finally, specific research gaps and needs related to plume dynamics, atmospheric transport and deposition processes, and the atmospheric chemistry of wildland fire emissions are identified and discussed.

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1. Introduction

Greenhouse gases and aerosols emitted from wildland fires are transported away from burning areas due to local atmospheric circulations induced by the fires and ambient atmospheric circulations. The lofting of smoke plumes from wildland fires through plume rise processes to higher levels in the atmosphere is generally followed by the horizontal transport of those plumes to locations far removed from the ignition source of wildland fires. During the vertical rise and the horizontal transport of plumes

through the atmosphere, many of the chemical and gaseous species that comprise wildland fire smoke, including greenhouse gases, undergo chemical reactions. These chemical reactions can further affect plume concentrations and distributions of greenhouse gases and aerosols and their ultimate impact on the climate system through radiative forcing. This paper provides an overview and summary of the current state of knowledge regarding critical atmospheric transport and chemistry processes that affect the distribution and concentrations of greenhouse gases and aerosols emitted from wildland fires or produced through subsequent chemical reactions in the atmosphere. Current modeling tools and monitoring techniques used for assessing wildland fire plume behavior are also discussed. Finally, suggestions for future research are provided to further advance our understanding of the critical atmospheric processes involved in wildland fire plume dynamics.

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2. Plume rise processes

Smoke plume-rise height is characterized as the maximum height a smoke plume can reach vertically in the atmosphere. For a well-developed plume, it is measured at the point where a smoke plume bends from vertical rising to horizontal transport. Typical smoke plume-rise heights range from hundreds of meters for prescribed fires to thousands of meters for wildfires, with occasional stratospheric penetration occurring for the most energetic fires (Gabbert, 2010). Plume rise is an important factor for local and regional air quality. Fire emissions injected at higher elevations are likely to be transported out of the local burn site and may affect air quality in downwind locations, including sensitive populations and urbanized areas. Furthermore, heat, water, and particles (including black carbon and other aerosols) emitted from fires impact atmospheric thermal, dynamical, and hydrological conditions and processes (Liu et al., 2014). Plume rise and the vertical distribution of gases and particles within smoke plumes are critical factors in assessing the downwind impacts of smoke plumes from wildland fires.

2.1. Modeling tools

Plume height is a parameter required by many regional air quality models. The Community Multiscale Air Quality (CMAQ) model and software suite (Byun and Ching, 1999; Byun and Schere, 2006) incorporates the Sparse Matrix Operator Kernel Emissions Modeling System (SMOKE) (Houyoux et al., 2002) to provide plume rise as part of initial and boundary conditions for elevated emission sources. In an early smoke model version of CMAQ and SMOKE, the Briggs scheme (Briggs, 1975) originally developed for power plant stacks was used to calculate fire smoke plume rise. The Briggs scheme is a two-thirds law integral model based on differential equations governing fluxes of mass, momentum, and energy through a plume cross section. Plume rise is calculated from both emission properties, such as initial buoyancy flux and exit velocity, and ambient properties, such as wind and thermal stability. The performance of the Briggs scheme is dependent on the relative importance of these properties. Performance is enhanced if thermal turbulence generated by the plume buoyancy dominates over mechanical turbulence generated by the ambient airflow. The typical plume rise scenarios are power plant stacks and smoke plumes in relatively calm wind conditions. Thus, in theory, if the plume momentum flux dominates, the Briggs scheme may not perform well. Guldberg (1975) compared the accuracy of the Briggs scheme with two other schemes in modeling the heights of hot, buoyant plumes and found that the Briggs scheme best predicted the observed plume heights during periods of low wind speed.

Analysis of the valid applications of the Briggs scheme emphasizes the importance of developing smoke plume rise schemes specifically for wildland fires. A number of wildland fire smoke plume rise models have been recently developed. These models can be divided into three types (Empirical, Dynamic, and Hybrid models). Empirical models are developed based on field and laboratory measurements and analyses using statistical methods or similarity theory. Expert opinion is used in some models. Empirical models are algebraic expressions that require no time or space integration. Because of simplicity, empirical models are easily used by fire and air quality managers. Harrison and Hardy (2002) developed an empirical model to estimate plume rise using peak flame power based on the measurements of a large number of prescribed burns in the northwestern US. The Briggs scheme was modified in the Fire Emission Production Simulator (FEPS) scheme by converting the heat flux from a fire to a buoyancy flux (Anderson et al., 2004). The Western Regional Air Partnership (WRAP, 2005) used

a climatological method by specifying predefined plume bottom and plume top and a predefined diurnal temporal profile for each fire.

Dynamical models are the second type of wildland fire smoke plume rise models. They consist of differential equations governing fluxes of mass, momentum and energy with solutions found through time and/or space integration. Because of their complexity, dynamical models are usually employed as research tools. One example is the dynamical model (Freitas et al., 2006) based on a one-dimensional dynamic entrainment plume model (Latham, 1994). An extended set of equations, including the horizontal motion of the plume and the additional increase of the plume size, are solved to explicitly simulate the time evolution of the plume rise and determine the final injection layer. Dynamical models, specifically high resolution atmospheric boundary-layer models and large-eddy-simulation models that are able to resolve atmospheric circulations and thermodynamics within forest vegetation layers, are now being applied to prescribed fire episodes in order to assess the impacts of forest vegetation on initial plume rise and local smoke dispersion. For example, Kiefer et al. (2011a, 2011b) developed a canopy sub-model for the Advanced Regional Prediction System (ARPS) (Xue et al., 2000, 2003), and then used ARPS to simulate the effects of forest vegetation on the atmospheric boundary-layer dynamics that influenced the initial plume rise from a prescribed fire in the New Jersey Pine Barrens. Results from their work suggest that forest vegetation has a significant impact on atmospheric turbulence and the resulting vertical and horizontal dispersion of wildland fire smoke emissions in the lower atmospheric boundary layer.

The third type is a hybrid of the empirical and dynamic smoke models. One example is Daysmoke (Achtemeier et al., 2011), which simulates smoke particle movements using statistical and stochastic relations. Daysmoke consists of four sub-models: an entraining turret model, a detraining particle model, a large eddy parameterization for the mixed boundary layer, and a relative emissions model that describes the emission history of the prescribed burn. These relations appear in differential equations and therefore require time and space integration. A rising smoke plume is described by a train of rising turrets of heated air that sweep out a three-dimensional volume defined by plume boundaries expanding with time through entrainment of surrounding air through the sides and bottoms as they ascend. Daysmoke was developed specifically for prescribed burning and is an extension of ASHFALL, a model developed to simulate deposition of ash from sugar cane fires (Achtemeier, 1998). In comparison with wildfires, the role of buoyancy generated by prescribed fires often is relatively smaller because of the smaller amount of heat released. Mechanical turbulence in the boundary layer is usually more important than the buoyancy associated with prescribed fires in governing how prescribed fire smoke plumes behave in the boundary layer.

Smoke transport and dispersion models are being used operationally on prescribed burns and wildfires to simulate and predict the transport and dispersion of smoke and to estimate particulate matter concentration at ground level. These modeling tools fall into several categories that are described and summarized in Goodrick et al. (2012): box models, Gaussian plume models, Lagrangian puff and particle models, Eulerian grid models, full physics models, and smoke modeling frameworks. The complexity of these models range from a simple box representing an airshed with a defined top of the mixing layer and horizontal dimensions defined by the spatial extent of the wildland fire airshed, to more complex modeling frameworks that link individual fuel loading, fuel consumption, emissions, and smoke trajectory and concentration models in a modular framework to predict real-time smoke trajectories and concentrations.

2.2. Measurements

Smoke plume-rise measurements are essential for model development and evaluation. The lack of systematic smoke plume rise measurements has been a big challenge for modelers. Three basic techniques have been used for determining the height of smoke plumes: plume sampling by aircraft, photographing the plume, and most recently, using remote sensors. Some substantial progress has been made in applications of existing and new remote sensing techniques. Melnikov et al. (2008), Jones and Christopher (2008), and Tsai et al. (2009) applied weather radars, such as the Weather Surveillance Radar-1988 Doppler (WSR-88D), that are part of the U.S. national operational weather radar network to detect smoke plume reflectivity and structure. LIDAR (Light Detection and Ranging) is another optical remote sensing technique extensively used for smoke detection (e.g., Mikkelsen et al., 2002; Lavrov et al., 2003; Colarco et al., 2004; Kovalev et al., 2009; Wold et al., 2010; Liu et al., 2012). LIDAR emits a laser beam and receives backscatter signals from particles, such as those found in smoke plumes. LIDAR has the advantages of equipment mobility, robustness, and low energy consumption and can be placed on ground or aboard aircraft and satellite (Kovalev and Eichinger, 2004; Fujii and Fukuchi, 2005; Kovalev et al., 2009). Satellite techniques for smoke plume height detection include the Cloud-Aerosol LIDAR with Orthogonal Polarization (CALIOP) aboard the Cloud-Aerosol LIDAR and Infrared Pathfinder Satellite Observations (CALIPSO) satellite (Winker et al., 2006) and the Multi-angle Imaging SpectroRadiometer (MISR) aboard the NASA TERRA satellite. Many applications of satellite techniques for smoke plume detection and analysis have been reported recently, including Labonne et al. (2007), Kahn et al. (2007, 2008), Diner et al. (2008), Raffuse et al. (2009), and Amiridis et al. (2010). The satellite techniques offer global coverage but at temporal frequency as low as every 15 days passing over a specific location.

Available measurements provide useful data for model evaluations. Raffuse et al. (2009) compared plume rise of wildland fires across the continental US between the simulation with the FEPS scheme and measurements from the MISR satellite remote sensing. It was found that the simulated smoke plume rise was systematically lower for weak fires and higher for large fires. Smoke plume height was recently measured using a ground-based ceilometer for 20 prescribed burns in the southeastern US (Liu et al., 2012). About half of the burns had burned areas exceeding 404.7 ha (1000 acres). Average smoke plume height was approximately 1 km, with plume heights trending upward from winter to summer (Fig. 1). These results could be used as an empirical guideline for fire managers to estimate smoke plume height in the southeastern US when modeling and measurements are not available. The average could be used as a first-order approximation, and a second-order approximation could be obtained by using the average for spring and fall seasons, and decreasing (increasing) by 0.2 km the average for the winter (summer) season. Additional knowledge of the fuel types and moisture conditions present could further improve the estimates made using the above guidelines.

LIDAR allows continuous monitoring of smoke-polluted atmospheres adjacent to severe wildfires in real time, providing temporal and spatial variation of aerosol properties, plume heights, plume dynamics, the direction and rate of smoke plume movement; and smoke layering at 2–4 km heights, typical in smoke-polluted atmospheres when the morning inversion positions the smoke in a layer located above the planetary layer (Kovalev and Hao, 2008). The scanning capabilities of the LIDAR can be categorized as follows (Kovalev et al., 2008):

- Two-dimensional spatial scan: An image (“slice”) of relative smoke concentration is developed from individual LIDAR lines of sight (horizontal or vertical).

- Three-dimensional spatial scan: A series of horizontal scans at different elevation angles in a given region of the atmosphere.
- Time-domain scan: Data collected from a single line of sight (general in zenith) for any particular time duration.

Examples of the application of LIDAR technology for measuring wildland fire plume characteristics are provided by Kovalev et al. (2008, 2011a, 2011b), and Urbanski et al. (2010) who used the USDA Forest Service's Fire Sciences Laboratory (FSL, Missoula, MT) mobile scanning LIDAR system to monitor five different plume types during multiple wildfire events:

1. Dense fire plumes concentrated over large wildfires and in their near vicinity (Tripod Complex Fire, Winthrop, Washington, August, 2006);
2. Spotted local fire plumes scattered within a wildfire area (Bull Fire, New Mexico, June–July, 2005);
3. Well-defined and stable multiple smoke horizontal layering, located mostly at the heights of ~2000–3000 m with relatively clear air below and above the layers (I-90 Fire, Montana, August, 2005);
4. Downwind smoke plume that bends from vertical uprising to horizontal or close to horizontal transport (Kootenai Creek Fire, Montana, July–August 2009);
5. Highly dispersed smoke haze far downwind of large fires (Tripod Complex Fire, Okanogan, Washington, August 2006).

Defining the upper boundary height of the region of increased backscatter as the maximal height where the aerosol heterogeneity is detectable, as proposed by Kovalev et al. (2011a), provides the maximal sensitivity for the smoke plume detection in the unavoidable presence of noise in the LIDAR signal. A special data processing methodology based on this definition has been developed for extracting information about the plume heights and their spatial and temporal changes in smoke polluted atmospheres.

For the Kootenai Creek Fire, which burned 2000 ha of conifer forest from mid-July to early September, a combined LIDAR-aircraft smoke plume investigation was performed using the FSL LIDAR and a Forest Service Region 1 Aircraft. The LIDAR and the airborne measurement data obtained in the area of the Kootenai Creek Fire on August 27, 2009, are shown in Figs. 2 and 3. The LIDAR measured smoke plume heights and plume profiles show good agreement with the maximal smoke plume height and PM_{2.5} concentration profiles determined from the aircraft instrumentation.

These monitoring studies suggest that mobile scanning LIDAR may be the most appropriate tool for ground-based monitoring of wildfire smoke-plume dynamics and heights at different distances from active fires. It was successfully used for real-time determination of smoke plume dispersion, plume top heights, and spatial boundaries.

2.3. Applications

Measurements and modeling of smoke plume rise have been used to improve our understanding of smoke plume rise properties. One of the important smoke properties for plume rise identified recently is plume updraft core number (Achtmeier et al., 2004). A single smoke plume may consist of several updraft cores, resulted from multiple ignitions at different locations within a burning site, smoke interactions, and other processes. Multiple-core updrafts are not as efficient as are one-core updrafts in the vertical transport of equivalent amounts of smoke mass. Multiple-core updrafts have smaller initial updraft vertical velocities and temperature anomalies and thus decreased initial buoyancy. Furthermore, entrainment more effectively reduces buoyancy of

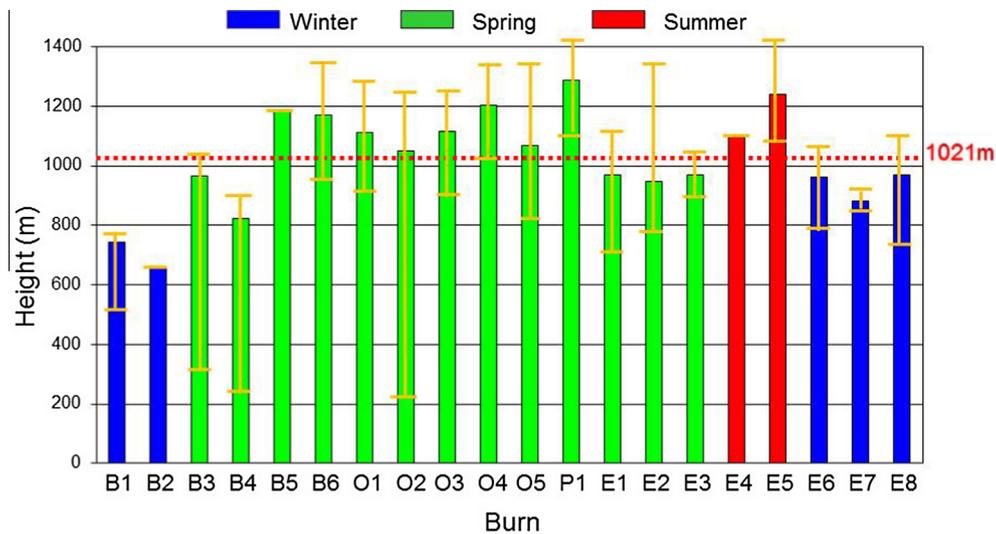


Fig. 1. Smoke plume height means (bars) and ranges between minimum and maximum hourly values (yellow lines) for each burn. The dotted line is average smoke plume height of all burns. Burns in different seasons are distinguished by colors. (From Liu et al., 2012.)

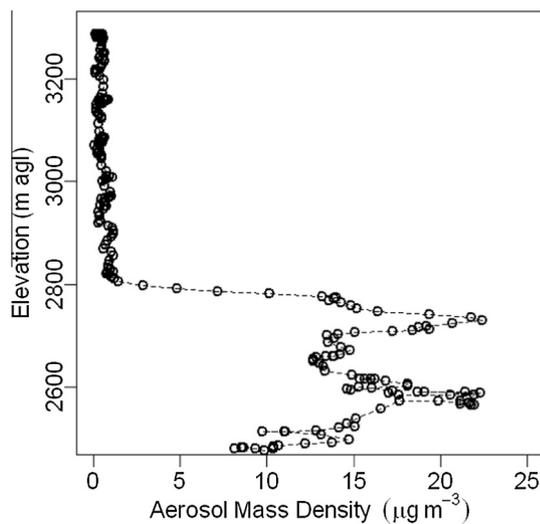


Fig. 2. Vertical profile of aerosol mass concentration measured ~10 km downwind of the Kootenai Creek Fire at 1600 LT on August 27, 2009. The vertical profile clearly identifies the top of the smoke plume located ~2790 m. (From Urbanski et al., 2010.)

multiple core updrafts with smaller effective plume diameters. In comparison with one-core plume updrafts, multiple-core updrafts lack vigor in transporting smoke into the free atmosphere above the mixing layer. This property was found to be the most important parameter along with entrainment rate for prescribed burning smoke plume rise simulated with Daysmoke (Liu et al., 2010).

Smoke plume rise models have been incorporated into fire and air quality modeling systems to provide smoke plume rise and vertical profiles for air quality simulation and prediction of fire emissions. For example, the FEPS and WRAP schemes are parts of the Bluesky framework that provides fire emission information to CMAQ (Pouliot et al., 2005; Larkin et al., 2009; Strand et al., 2012). Daysmoke has been incorporated into the SHRMC-4S framework (Liu et al., 2009) for simulating smoke and the air quality effects of prescribed burning and into the adapted-grid CMAQ to simulate sub-grid smoke transport and plume rise (Garcia-Mendez et al., 2010). Applications of smoke plume rise models have provided evidence for the importance of smoke plume rise to

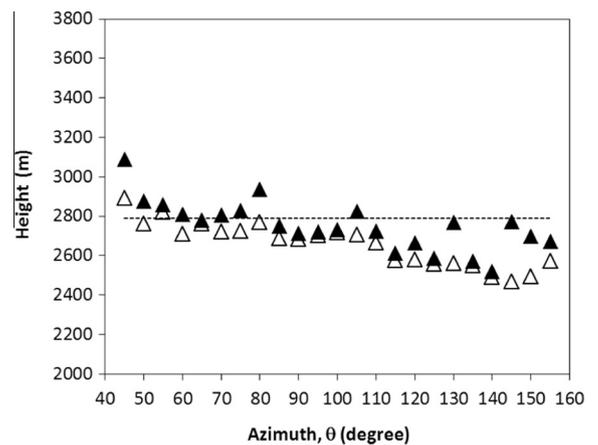


Fig. 3. Maximum heights of the smoke plume determined at different azimuthal directions measured in the vicinity of the Kootenai Creek Fire on August 27, 2009, during the period from 12:09 to 12:27 LT. The horizontal dashed line indicates the smoke plume height determined from airborne measurements. (From Kovalev et al., 2011a.)

simulation of the air quality impacts of wildfires (Liu et al., 2008). There is also evidence that inclusion of smoke plume rise modeling can improve regional air quality modeling (e.g., Liu et al., 2010). Smoke plume rise models have also been applied to smoke-atmosphere interaction studies. The dynamic model developed by Freitas et al. (2006) was coupled with the Weather Research and Forecast (WRF)-Fire modeling system (Mandel et al., 2011) and used to simulate the intense wildfires during the 2004 Alaska fire season. The simulation indicated a strong impact of fire emissions on clouds and cloud microphysics (Grell et al., 2011).

3. Atmospheric chemistry of emissions

This section focuses on the atmospheric fate of non-carbon dioxide (CO_2) gas-phase constituents and the relationship of their atmospheric chemistry to radiative forcing. Trace gases impact radiative forcing by changing atmospheric composition – the concentration of gases and aerosols spatially and temporally as well as aerosol properties (phase, shape, chemistry, and optical

properties). The impact of fire emissions on atmospheric composition and the realized radiative forcing depends on the composition of the emissions, location, and ambient environment (chemical and meteorological). While CO₂ and water vapor dominate emissions from wildland fires, smoke is nonetheless a rich and complex mixture of gases and aerosols (van der Werf et al., 2010; Wiedinmyer et al., 2011). The contribution of biomass burning to emissions of carbon to the atmosphere was identified as an important source of radiatively and photochemically reactive trace gases in 1980 (Seiler and Crutzen, 1980). A previous study (Crutzen et al., 1979) had investigated the atmospheric budgets of trace gases in the atmosphere including carbon monoxide (CO), molecular hydrogen (H₂), nitrous oxide (N₂O), nitric oxide (NO), nitrogen dioxide (NO₂), and carbonyl sulfide (COS). Seiler and Crutzen (1980) showed that these trace gases were emitted into the atmosphere in large quantities by measuring emission rates of trace gases relative to CO₂ in the smoke plumes of forest and grassland wildland fires. CO₂ is relatively inert and it is the more reactive, if less abundant species that are responsible for much of the important atmospheric chemistry associated with fire emissions. The balance of emissions, in order of decreasing quantity, are CO, methane (CH₄), non-methane organic compounds (NMOC), aerosols (organic and black carbon), nitrogen oxides (NO_x), ammonia (NH₃), and limited quantities of other trace gases (e.g., sulfur dioxide (SO₂), N₂O) and hydrogen chloride (HCl). While CO₂, CH₄, and aerosols produce direct radiative forcing, the non-CO₂ gases also affect radiative forcing through their photochemical processing, which impacts levels of CO₂, CH₄, tropospheric ozone (O₃), stratospheric water vapor, and aerosols (Shindell et al., 2009).

Gases and aerosols are ultimately removed from the atmosphere and transported to the Earth's surface by wet or dry deposition. Wet deposition refers to the scavenging of gases and aerosols by atmospheric hydrometeors (clouds, fog droplets, rain, and snow) and subsequent delivery to the surface. The transport of gases and aerosols to the Earth's surface in the absence of precipitation is referred to as dry deposition. A detailed description of wet and dry deposition may be found in Seinfeld and Pandis (2006). Prior to delivery to the Earth's surface, atmospheric trace gases, regardless of source (wildland fire, biogenic emissions, or anthropogenic activities), may undergo chemical reactions or physical transformation. The gas-phase chemistry of the troposphere oxidizes CO, CH₄, and NMOC, eventually converting the carbon to CO₂, unless dry or wet deposition preempts the process. The gas-phase degradation of CO and CH₄ is initiated by reaction with hydroxyl (OH) radicals; in addition to reaction with OH, the oxidation of NMOC may be started by reaction with O₃, nitrate (NO₃) radicals, or photolysis by solar radiation. Because the reactions of NMOC and CO with OH are much more rapid than the CH₄ + OH reaction, increases in CO and NMOC levels reduce the availability of OH for CH₄ oxidation and thereby increase CH₄ concentrations and radiative forcing. NO_x and NMOC serve as precursors to the formation of O₃ and secondary organic aerosol (SOA). In the presence of NO_x, the products of the initial NMOC reactions can lead to the production of O₃ and SOA (Hallquist et al., 2009; Jaffe and Wigder, 2012). CH₄ and CO oxidation may also produce O₃ (see Seinfeld and Pandis, 2006, for an overview of tropospheric chemistry). SOA are formed when intermediates and products of NMOC oxidation are involved in gas-particle conversion processes such as nucleation, condensation and heterogeneous, and multiphase chemical reaction (Hallquist et al., 2009). The production of O₃ and SOA depends on the mix of NMOC, the initiating chemical reactions, and ambient conditions (e.g., levels of NO_x, relative humidity, solar radiation, temperature, and pre-existing aerosols (Hallquist et al., 2009; Jaffe and Wigder, 2012).

Wildland fires release substantial amounts of O₃ and SOA precursors (Akagi et al., 2011; van der Werf et al., 2010; Wiedinmyer

et al., 2011). The production of O₃ and SOA precursors varies with fuel type and combustion conditions. Combustion efficiency impacts the levels of NMOC and NO_x. Low combustion efficiency enhances NMOC emissions and reduces NO_x emissions. In addition to combustion efficiency, NMOC production also varies with fuel type. Unlike fossil fuel combustion, NO_x emitted from fires is derived almost exclusively from nitrogen contained in the biomass burned; therefore fuel chemistry is an important factor in NO_x production (Burling et al., 2010). In addition to the variability of emissions, production of O₃ and SOA depends on the mix of NMOC, the initiating chemical reactions, and ambient conditions such as levels of NO_x, relative humidity, solar radiation, temperature, and pre-existing aerosols (Hallquist et al., 2009; Jaffe and Wigder, 2012). Therefore, the impact of fires on O₃ and SOA is highly variable.

Globally, 75% of the total organic aerosol (OA) flux to the atmosphere is SOA (Hallquist et al., 2009). The oxidation of NMOC emitted from terrestrial vegetation is the dominant global source of OA, accounting for 60% of production (Hallquist et al., 2009). Given the variability of emissions and the importance of ambient conditions (such as levels of NO_x, relative humidity, solar radiation, temperature, and pre-existing aerosols), the production of SOA attributable to fires is highly uncertain. Complicating matters is the fact that a large fraction of primary OA emissions in concentrated plumes (e.g., near the fire) may evaporate as the plume is diluted to ambient conditions (Hallquist et al., 2009). The evaporated vapors may then be oxidized to lower volatility compounds that recondense to aerosol. For example, in a fresh biomass fire plume Akagi et al. (2012) observed an initial decrease in OA followed by a slow increase after about 1.5–2 h. Globally, fire generated SOA are estimated to account for ~10% of OA and ~20% if recycling of gases evaporated from primary aerosol to secondary aerosol is considered (Hallquist et al., 2009). By comparison, primary OA from fires, excluding the fraction that is thought to evaporate, are estimated as <10% of the global OA source (Hallquist et al., 2009).

A large number of atmospheric observations, covering the tropical, subtropical, boreal, and temperate regions have shown O₃ enhancement in aged smoke plumes (Jaffe and Wigder, 2012). A few observations of plumes from boreal fires have shown slight depletion in O₃. In general, aged smoke plumes show O₃ enhancement, with the enhancement in tropical/subtropical wildland fire smoke plumes being higher than that observed in boreal/temperate wildland fire plumes. In a recent review of O₃ production from biomass burning, Jaffe and Wigder (2012) found average O₃ enhancements in tropical/subtropical smoke plumes were three times the enhancement for boreal/temperate plumes. While a host of factors influence O₃ in smoke plumes, greater flux of solar radiation and higher temperatures contribute to the greater O₃ enhancement observed in the tropics and subtropics. Jaffe and Wigder (2012) estimate global O₃ from biomass burning of 175 Tg yr⁻¹. This is only 3–5% of total chemical O₃ production estimated in global model budgets of tropospheric O₃ (Wu et al., 2007). However, because wildland fires tend to be concentrated spatially and temporally, emissions can have a significant influence locally and regionally on O₃ during periods of intense burning. While the radiative forcing due to biomass burning O₃ is small in comparison to aerosol forcing (Jeong et al., 2008; Pfister et al., 2008), wildfires have been shown to significantly elevate O₃ levels in US metropolitan areas (Jaffe et al., 2008; Morris et al., 2006; Pfister et al., 2008).

4. Long-range tropospheric transport

Greenhouse gases and aerosols emitted from wildland fires or produced through chemical reactions involving gaseous species

contained in smoke plumes can be transported to locations far removed from where fires are occurring. Once these gases and aerosols are lofted into the mid to upper troposphere through plume rise processes and deep convection that commonly occurs in the tropical and mid-latitude regions, large-scale circulation systems in the troposphere may be able to transport them to other regions and continents where they can affect air quality and atmospheric and surface radiative processes. The body of scientific literature devoted to describing and analyzing actual atmospheric transport events associated with wildland fire emissions is significant. Some of the key published studies are described below.

One of the most comprehensive studies on long-range atmospheric transport processes associated with atmospheric pollutants in general is described by the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) (TF HTAP, 2010). This report provides clear observational evidence that Northern Hemispheric concentrations of ozone and particulate matter (including black carbon), some of which are associated with ozone precursor and particulate matter emissions from wildland fires, are influenced by long range intercontinental and hemispheric transport. The report indicates westerly winds in the mid-latitude troposphere are responsible for the relatively rapid transport of pollutants from North America to Europe, from Europe to the Arctic and central Asia, and from East Asia to North America. The TF HTAP study found that this relatively rapid (<2 weeks), long range, mid and upper tropospheric transport often occurs within the warm conveyor belt of mid-latitude cyclones. Intercontinental transport within the lower troposphere also occurs but is much slower. The Photochemical Ozone Budget of the Eastern North Pacific Atmosphere (PHOBEA) research campaign found evidence of Siberian boreal fire emissions that were transported to the northwestern coast of Washington State during the spring of 2002 (Bertschi et al., 2004). Using satellite data, Damoah et al. (2004) found that emissions from severe forest fires in southeast Russia in May 2003 were transported eastward across Alaska, Canada, Europe, and back to Russia in the span of 17 days. The observational evidence of long-range intercontinental transport is also supported by modeling studies, such as Stohl et al. (2003a) who used a Lagrangian particle dispersion model (FLEXPART) to show that anthropogenic sources and forest fire emissions can contribute to an accumulation of pollutants in the North American boundary layer (e.g., over California, Texas, and Florida) that can then be lofted into the mid and upper troposphere and transported rapidly to Europe.

In addition to the general west-to-east long-range transport of atmospheric pollutants that occurs across the globe, there is also a northward transport of pollutants to the Arctic region that can occur (Larkin et al., in press). Shindell et al. (2008) carried out a multi-model assessment of pollution transport to the Arctic region and found that the deposition of black carbon onto Greenland is most sensitive to North American emissions. Considering both European and North American black carbon emissions, roughly 40% of the total black carbon deposited on Greenland is transported from emissions sources in these continents. For the entire Arctic region however, the TF HTAP (2010) report indicated that the transport of North American black carbon emissions to the Arctic only accounted for an average of 5% of the surface black carbon observed there. For example, Sharma et al. (2006) found that black carbon measured in northern Alaska (Barrow) from 1989 to 2003 was mainly transported there from emissions sites in Russia/Siberia, with the induced large-scale circulations associated with the North Atlantic Oscillation playing a major role in the transport process. The modeling study of Koch and Hansen (2005) suggested that most of the black carbon in the present-day Arctic comes from industrial and biofuel sources in south Asia and from biomass burning in Russia. This finding was reinforced by Warneke et al.

(2009) who used observational data collected during the Aerosol, Radiation, and Cloud Processes affecting Arctic Climate (ARCPAC) field experiment and transport modeling results from the Lagrangian particle dispersion model FLEXPART to show that biomass burning in Siberia and Kazakhstan contributed to haze over the Alaskan Arctic in 2008.

Long-range tropospheric transport of gases and aerosols occurs at regional and continental scales as well. For example, smoke plumes originating from wildland fires in the Quebec region of Canada in 2002 and transported to the northeastern region of the US degraded atmospheric boundary layer (ABL) air quality and decreased solar radiation and air temperatures in the Baltimore, Maryland, and Washington DC areas (Colarco et al., 2004; Pahlow et al., 2005). The reduced air quality in these areas was the result of downward sweeps of smoke-laden air from the free atmosphere into the ABL. Sapkota et al. (2005) also examined the impacts of the 2002 Quebec fires on air quality in Baltimore and found that the long-range transport and downward mixing of the smoke plume into the ABL even resulted in the degradation of indoor air quality in Baltimore. Duck et al. (2007) observed the eastward transport of wildland fire emissions from Alaska and the Yukon Territory during the summer of 2004 to Nova Scotia, Canada. As part of the Intercontinental Chemical Transport Experiment – North America (INTEX-NA), Fuelberg et al. (2007) found that persistent high pressure systems over Alaska during the summer of 2004 provided ideal conditions for wildfires and the transport of fire emissions to distant locations such as southern Texas, Louisiana, and the Labrador Sea off the coast of Newfoundland, Canada. Smoke plumes originating from Alaskan and western Canadian forest fires and transported southward and eastward were found to exacerbate ozone pollution over Houston, Texas, in 2004 (Morris et al., 2006) via enhanced ozone production from chemical reactions involving ozone, nitrogen oxides, and hydrocarbons in the transported smoke plumes. Smoke aerosols from California forest fires in October 2003 were measured via the ICESat satellite and were found to have been transported to the northeastern US (Hoff et al., 2005). Aerosols emitted from California and Oregon forest fires during the summer of 2008 were measured over southwestern British Columbia using ground-based and satellite instruments (McKendry et al., 2011). Wotawa and Trainer (2000) were able to show that episodes of high CO concentrations in the southeastern US during the summer of 1995 were the result of long-range transport of smoke plumes from large forest fires in northern Canada. These smoke plumes also contained high concentrations of ozone, aerosols, and volatile organic compounds. Episodes of smoke transport to the southeastern US from wildland fires in Central America during the springs of 1998 and 2003 were also highlighted in observational and atmospheric mesoscale modeling studies carried out by Tanner et al. (2001) and Wang et al. (2006), respectively.

5. Stratosphere–troposphere exchange

Greenhouse gases and aerosols emitted from wildland fires or produced through chemical reactions involving gaseous species contained in smoke plumes have the potential for being transported vertically to the top of the troposphere where mixing into the stratosphere is possible. Gases and aerosols that are transported into the upper troposphere and lower stratosphere have the potential for altering the radiation balance and atmospheric chemistry occurring there, including chemical reactions that affect stratospheric ozone concentrations (Cofer et al., 1996; Waibel et al., 1999; Jost et al., 2004). Several studies focused on the transport of gases and aerosols, including smoke from forest fires, between the troposphere and stratosphere have been carried out over the last decade.

Separating the troposphere from the stratosphere is the tropopause. The World Meteorological Organization (WMO) has defined the tropopause as the lowest level at which the temperature lapse rate decreases to 2 K km^{-1} or less, and the lapse rate averaged between this level and any level within the next 2 km does not exceed 2 K km^{-1} (Stohl et al., 2003b; World Meteorological Organization, 1986; Hoinka, 1997). The height of the tropopause varies from the tropics to the poles. In the tropics, the tropopause is roughly 15–18 km above the surface. It slopes downward toward the poles, where its height is as low as 6–8 km above the surface (Holton et al., 1995). Although the temperature lapse rate in the stratosphere tends to inhibit the mass movement of air between the troposphere and stratosphere, mixing between the two atmospheric layers does occur. As described in Stohl et al. (2003b), on a global scale and over long time scales there is generally an upward transport of air from the troposphere to the stratosphere in the tropics (Plumb, 1996; Mote et al., 1996), a transport of air through the stratosphere from the tropics to the extratropics (Vaughn, 1996), and downward transport of air from the stratosphere to the troposphere in the mid- and upper-latitudes (Holton et al., 1995). On shorter spatial and temporal scales, however, stratosphere–troposphere exchange in the mid-latitudes is actually very episodic in nature and can occur in both directions (Stohl et al., 2003b). Evidence of mid-latitude transport of air from the lower troposphere to the lower stratosphere (lower stratospheric moist layers containing tracers from the Earth's surface) was actually reported by Poulida et al. (1996), Hintsä et al. (1998), Vaughan and Timmis (1998), and Ray et al. (1999). Stohl (2001) and Wernli and Bourqui (2002) pointed out that the rapid transfer of lower tropospheric air to the lower stratosphere is frequently associated with strong diabatic heating by latent heat release in the warm sector of extratropical cyclones.

Episodes of stratosphere–troposphere exchange in the mid-latitudes can result in the transport of wildland fire smoke from the troposphere to the stratosphere. Fromm et al. (2000) suggested a possible link between smoke from Canadian boreal forest fires and increased aerosol concentrations in the stratosphere in 1998, with significant convection in the atmosphere playing a critical role in the transport of aerosols into the stratosphere. Fromm and Servranckx (2003) followed up this initial study with a comprehensive case study of smoke transport from the Chisolm Fire in 2001 that occurred 160 km north of Edmonton, Alberta, Canada. Using fire plume data and imagery during the Chisolm Fire from NASA's Earth Probe Total Ozone Mapping Spectrometer (EP-TOMS), the Sea-viewing Wide Field of View Sensor (SeaWiFS), and the Advanced Very-High Resolution Radiometer (AVHRR), they concluded that under certain atmospheric conditions, extreme convection associated with forest fires and the ambient atmosphere can lead to the transport of smoke plumes from the planetary boundary layer to the upper troposphere and lower stratosphere. The actual troposphere to stratosphere transport of material was attributed to simple atmospheric advection and gravity wave breaking. Fromm and Servranckx (2003) further concluded in their study that this type of stratosphere–troposphere exchange occurs with some regularity at mid to high latitudes and that the radiative, chemical, and cloud microphysical impacts of the exchange may be significant.

Observations of mid-latitude boreal forest fire plumes occurring well into the stratosphere were also reported by Jost et al. (2004) as part of the Cirrus Regional Study of Tropical Anvils and Cirrus Layers – Florida Area Cirrus Experiment (CRYSTAL-FACE). Concentrations of CO as high as 193 ppb were observed in the stratosphere off the southwestern coast of Florida about 1.3 km above the local tropopause, whereas typical CO concentrations at that altitude are less than 50 ppb. Concentrations of CO₂, reactive nitrogen, and water vapor were also enhanced in this region. Using back-trajectory

analyses, Jost et al. (2004) determined that the sources of the enhanced CO off the southwestern coast of Florida were smoke plumes from wildland fires mainly in the Canadian province of Saskatchewan that were lofted into the stratosphere. They identified three possible contributors to the stratosphere–troposphere exchange: (1) convective system overshooting of its level of neutral buoyancy due to its inertia followed by mixing at the top of the convective system, (2) enhanced convection due to the additional heat and/or water vapor from the wildland fires, and (3) radiative self-heating and subsequent lofting of the injected plume.

6. Research gaps and needs

While the current state of knowledge regarding atmospheric processes involved in the transport and chemical make-up of smoke plumes associated with wildland fires is substantial, new modeling and observational research is still needed to address shortcomings in our understanding of fundamental fire–fuel–atmosphere interactions that govern the behavior of smoke plumes. Plume rise is determined by multiple factors, including fuel characteristics, fire behavior, emissions, canopy structure, fire-induced and ambient turbulence regimes, and basic atmospheric conditions. Building upon the successes of recent wildland fire experiments that have focused on fire–fuel–atmosphere interactions (e.g. Clements et al., 2007; Hiers et al., 2009; Seto and Clements, 2011; Seto et al., 2013), new comprehensive field measurements using *in situ*, upper-air, and downwind instrumentation of all these factors during wildland fire events are needed to continue to unravel the complexities of local and downwind plume behavior. Of particular importance are the roles of smoke-plume core number, forest-vegetation/boundary-layer-turbulence interactions, fire-induced latent and sensible heat fluxes, and the radiative forcing of smoke particles in affecting plume dynamics. A suite of observational datasets are also needed to compare and validate the performance of current and future smoke-plume dynamics models and smoke modeling frameworks, similar to the efforts of Tian et al. (2009), Achtemeier et al. (2011), Strand et al. (2012), and Raffuse et al. (2012).

For the current empirical plume-rise models used in the WRAP, Briggs, and FEPS schemes, new research is needed to evaluate the limitations of specifying hourly emissions and heat release via empirical profiles without distinction between prescribed fires and wildfires and without meteorological conditions considered. These characteristics of the WRAP, Briggs, and FEPS schemes potentially limit their capacity to simulate plume behavior for prescribed burns, which often last for only a few hours are much weaker than wildfires.

The most significant gap in our current knowledge of the atmospheric chemistry of wildland fire emissions, and biomass burning emissions in general, is their role in the formation of SOA. The importance of SOA formation from fire emissions has been observed to be highly variable, being significant under some circumstances, but unimportant in others. Narrowing this knowledge gap will require a better characterization of emissions and plume chemistry and an improved understanding of the influence of plume dynamics (rise, dilution, and cooling) and background chemical composition (e.g., urban or rural chemical environment).

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