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Particulate and trace gas emissions from prescribed burns in southeastern U.S. fuel types: Summary of a 5-year project ☆, ☆☆

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ABSTRACT

Management of smoke from prescribed fires requires knowledge of fuel quantity and the amount and composition of the smoke produced by the fire to minimize adverse impacts on human health. A five-year study produced new emissions information for more than 100 trace gases and particulate matter in smoke for fuel types found in the southern United States of America using state-of-the-art instrumentation in both laboratory and field experiments. Emission factors for flaming, smoldering, and residual smoldering were developed. Agreement between laboratory and field-derived emission factors was generally good in most cases. Reference spectra of over 50 wildland fire gas-phase smoke components were added to a publicly-available database to support identification via infrared spectroscopy. Fuel loading for the field experiments was similar to previously measured fuels. This article summarizes the results of a five-year study to better understand the composition of smoke during all phases of burning for such forests.

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1. Introduction

Prescribed burning is an accepted practice to manage biomass in the United States and throughout the world e.g. [1]. It is a particularly important practice in pine and eucalyptus forests, many of which have evolved in the presence of fire [2, 3]. A recent survey [4] reported that 2.62×10^6 ha of pine forest land in the southeastern U.S. (defined by the National Association of State Foresters) burned under prescribed conditions in 2011 for silvicultural purposes such as hazardous fuel reduction, wildlife habitat improvement, and forest regeneration. The survey reported 8.2×10^6 ha of forestry and agricultural lands burned by prescription in the conterminous U.S.; in the same year, wildfire burned 3.5×10^6 ha in the U.S. [5]. In 2004, estimated burned area worldwide was 3.2×10^8 ha of which temperate and boreal North America comprise approximately one percent [6]. The impact of emissions from prescribed burning worldwide are similarly a small percentage of total estimated biomass burning emissions [7, 8]; however, these emissions are regulated in many areas because they are planned.

Smoke is thus an important consequence of prescribed burning that must be managed [9]. A great deal of research has been performed since the 1970s characterizing the composition, production, and transport of smoke from such fires e.g. [9–13]. A recent review of the state of science behind estimation of the contribution of wildland fire to greenhouse gases and black carbon in the U.S. identified several areas of research that must be performed [14]. In particular, two areas of knowledge that still need improvement are (i) fuel characterization and smoke emissions, and (ii) the correlation(s) between the two [14]. This conclusion is likely true for many vegetation types throughout the world even though most experiments conducted over the past 30 years have been in the tropics and boreal forests. While many fuel types in the southeastern U.S. have been described for fire behavior and fire danger prediction e.g. [15–18], characterization of fuel bed components important for smoke production is more limited. Emissions characterization for both gases and particles is needed to determine potential impacts of prescribed burning on nutrient cycling, atmospheric albedo, human health, and visibility e.g. [13, 19, 20]. The southern U.S. was one of the first regions of the country to examine the impact of prescribed burning on air quality

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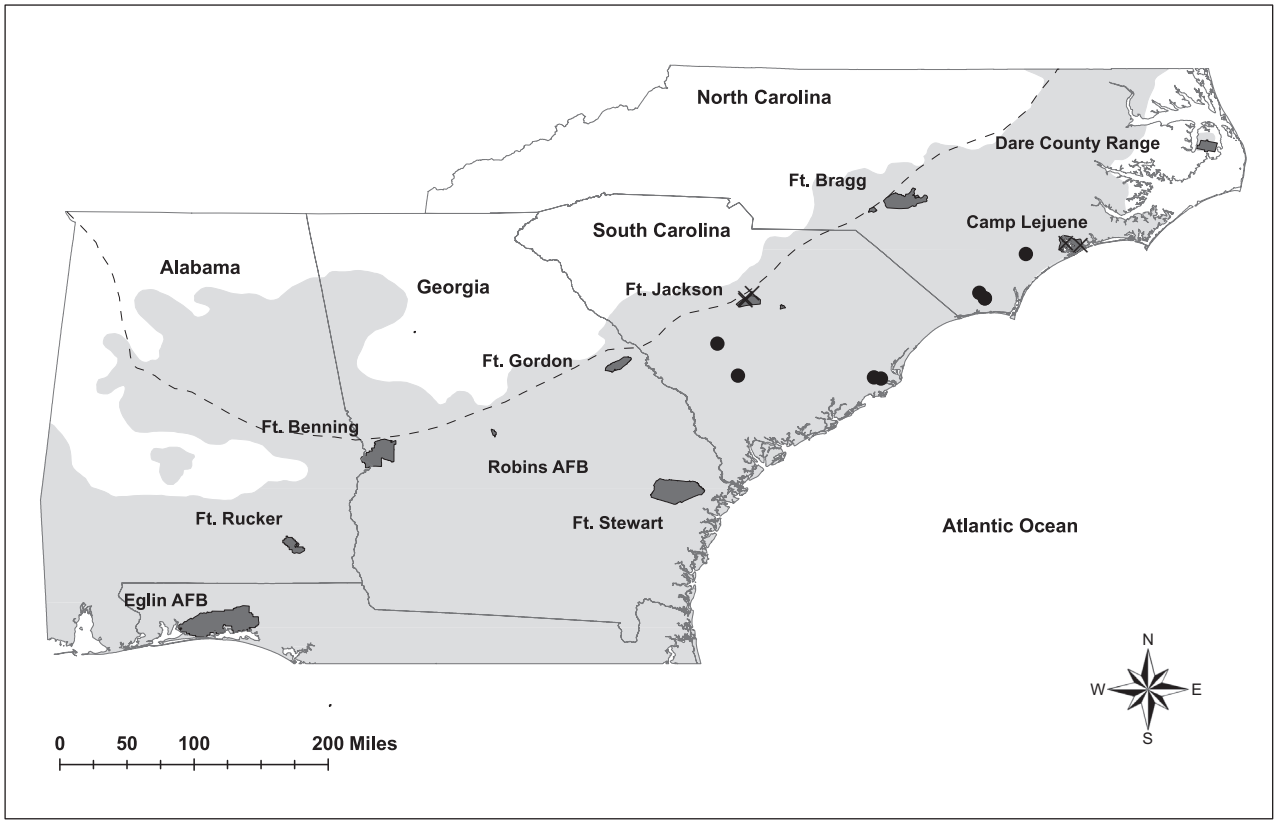

Fig. 1. Military installations (dark gray) containing fuel types measured as part of project. Light gray area denotes the estimated historical range of longleaf pine [95]. Dashed line indicates boundary of the coastal plain physiographic province ("fall line"). X denotes fires with airborne and ground samples, ● indicates airborne sample only. Longleaf range map available at http://www.auburn.edu/academic/forestry_wildlife/lpsdl/mappingfiles/LLP_Range_Little1971_Shapefile.zip.

in both the laboratory and in the field [9, 21]; recent advances in measurement devices provided an opportunity to improve knowledge of emissions using state-of the art instrumentation capable of real-time and *in situ* emissions measurements. Coupling well-controlled laboratory experiments using complex fuel beds with field measurements to determine applicability of laboratory results to actual conditions has seldom been accomplished.

The U.S. Department of Defense (DOD) manages much of its land in the southeastern U.S. to provide a wide range of ecological and economic benefits using ecological forestry [22]. Much of this land was historically dominated by a longleaf pine (*Pinus palustris* Mill.) forest maintained by frequent, natural, growing season fires [23]. Historically longleaf pine grew on approximately 3.7×10^7 ha and now occurs on less than three percent of its former range making it an ecosystem of concern [24, 25] (see light gray area in Fig. 1). Other southern pines (*P. taeda* L., *P. elliottii* Engelm., *P. serotina* Michx., *P. clausa* (Chapm. ex Engelm.) Vasey ex Sarg.) grow in this same range and rely on fire as well to maintain site dominance through light and resource competition control [2, 26]. Removal of fire from these ecosystems results in encroachment by hardwood trees and shrubs that threatens endangered species and limits military training [27]. Fuel accumulates in these forests faster than in other parts of the U.S. due to favorable growing conditions. As a consequence, as few as 4–6 fire-free years can cause a major fuel buildup making wildfires more difficult to control, and more damaging to the pine overstory when they do occur. The long-term impacts of such a change in the use of prescribed fire are currently unknown and a complex function of many factors [14, 28]. Longleaf pine grows in close proximity to many wetland areas which are also subject to wildland fire [26]. Pocosin is a dense shrub/pine ecosystem found in wetlands on deep organic soils that is extremely flammable during drought and often features intense fire behavior that can be problematic to suppress. Similar shrub-dominated ecosystems produce comparable fire behavior throughout the world [29]. Heavy smoke resulting from smoldering in the deep organic soils can be injurious to human health e.g. [30, 31]. Some areas with pocosin fuels and deep organic soils are managed by the U.S. Department of Defense, [32–34] such as the Dare County Range seen in Fig. 1.

In 2008 the Strategic Environmental Research and Development Program (SERDP) initiated a program to characterize the fuels, smoke chemistry and transport associated with prescribed burning on DOD bases in the U.S. Three related projects were completed and final reports are now available [35–37]. Detailed measurements of gaseous and particulate emissions were made in laboratory and field experiments. Here we summarize the work that has accurately measured the emissions of particles and numerous gases for the wildland fuels commonly found in the coastal plain and sand hills regions of the southeastern U.S. [26]. The many detailed results of these studies have largely been published in the refereed literature and are all contained in the final report [37]; this paper attempts to summarize the entire project in a succinct fashion, and in particular attempts to provide some key findings for scientists and users in the southern U.S. The results are also potentially applicable to many other similar pine and shrub-dominated ecosystems throughout the world. Emphasis is placed on the emission factors (EFs) derived for fires occurring in such ecosystems, as well as the relationships between the laboratory- and field-derived EFs.

2. Methodology

The project consisted of three components: (1) a laboratory effort to derive the infrared spectral absorption coefficients of

those gases contained in smoke from wildland fires and thus expand and improve the detection capability of infrared-based gas sampling instruments, (2) detailed measurements of both gaseous and particulate emissions from burning fuel beds in a large combustion laboratory, and importantly, (3) detailed ground and aerial measurement of gaseous and particulate emissions from operational prescribed burns at Camp Lejeune in North Carolina (coastal plain) and Fort Jackson in South Carolina (sand hills) (Table 1, Table 2).

2.1. Improved infrared instrumentation and spectral database

A useful technique for the measurement of gas-phase emissions from wildland fires is Fourier transform infrared [FTIR] spectroscopy. Compared to visible light, smoke is far more transparent to infrared light due to reduced Mie scattering at the longer IR wavelengths. Other advantages of FTIR include a nonintrusive measurement of a variety of flaming and smoldering phase compounds over an integrated path through the same air parcel. The unique spectral absorption features of the gaseous species measured means the technique is resistant to interference and can provide (with appropriate IR reference data) unambiguous compound identification. Moreover, FTIR has high temporal resolution for monitoring of dynamic processes related to emissions of biomass fires [22]. FTIR spectroscopy also provides the possibility to simultaneously measure a wide range of chemical species in smoke with good sensitivity (parts per billion levels in some cases) and high temporal resolution (1–2 s).

The FTIR instrument used in the laboratory burns in 2009 was improved by replacing the original Midac FTIR interferometer [38] with a Bruker IRcube spectrometer. The original Infrared Analysis, Inc. open-path White cell [39] was not changed. The improved Bruker IR Cube FTIR was later coupled to a closed-path White cell for the airborne measurements in 2011; the original Midac instrument was used for the airborne measurements in 2010 (see Section 2.3). Besides the FTIRs, a wide variety of instruments was used in the project to describe characteristics of the gas-phase and particulate emissions from the laboratory and field fires. These fires also provided the opportunity to compare performance of different instruments, techniques [40, 41] and performance of the same type of instrument at different sampling locations [42].

The PNNL infrared spectral library [43, 44] was originally developed for other purposes, and formerly contained about 400 compounds, of which perhaps 50 were associated with biomass burning. For this project, four criteria were used to select additional chemical species whose spectra should be added to the library: (1) the species had been measured in smoke by non-IR methods or was expected to occur based on known chemistry, (2) the species was expected to have reasonably strong IR absorption features in the 1300–700 cm^{-1} fingerprint region thus eliminating species such as Br_2 or elemental Hg, (3) the species had a vapor pressure $>\sim$0.01 Torr or a boiling point of $<$250–300 °C, so that it was both amenable to laboratory measurement and would not immediately condense in a cooling smoke plume and (4) the species must be sufficiently stable in an N_2 bath gas in the laboratory (for minutes to hours) so the measurement could actually be completed. Using vetted database protocols [43, 44] several dozen species were added to the spectral library as discussed below (Section 3.1).

2.2. Laboratory measurement of emissions

In the laboratory component, fuels representative of vegetation commonly managed by prescribed burning were collected and burned under controlled conditions at the U.S. Forest Service (USFS) Fire Sciences Laboratory (FSL) in Missoula, Montana. The fuels were burned under constant environmental conditions using techniques to promote near total consumption of the fuel under a chimney hood that captured all of the emissions produced. The emissions from these laboratory burns were analyzed with a suite of state-of-the-art instrumentation too large for collective field deployment on operational prescribed burns [37]. Seldom, if ever, has such a suite of instruments been assembled to measure biomass emissions.

At the FSL, gas-phase measurements were performed by (1) an open-path Fourier transform infrared (OP-FTIR) instrument described previously [Section 2.1], (2) two proton-transfer-reaction mass spectrometers (PTR-MS) for measurement of non-methane organic compounds (NMOC), (3) proton-transfer ion trap-mass spectrometry (PIT-MS) for NMOC, (4) negative-ion proton-transfer chemical-ionization mass spectrometry (NI-PT-CIMS) for detection of organic and inorganic acids, (5) gas chromatography mass spectrometry (GC–MS) for NMOC, (6) canister sampling followed with analysis by gas chromatography flame ionization detection (GC-FID) for low molecular weight hydrocarbons, (7) a LICOR CO_2 analyzer, and (8) TECO CO analyzer. Due to platform space and load restrictions, one PTR-MS and the GC–MS were housed in a control room adjacent to the combustion chamber and connected to the stack via a long Teflon sampling line. Together these instruments provided measurements of approximately 120 gas-phase components.

To characterize the particle emissions, several instruments located on the platform measured size distributions, particle number, mass, and chemical composition. Instrumentation included

Table 1
Objectives and key findings of study to improve emissions for prescribed burns in the southern United States.

Objective	Key accomplishments/Findings
Improved infrared instrumentation and spectral database	• Added 53 gases to spectral library to expand remote measurement of biomass burning emissions • More accurate estimation of emissions using FTIR due to increased sensitivity of instrument • Temporal measurement of 10 additional trace gases measured in the lab using FTIR • Field comparison of FTIR instruments indicate that measurement location (ground versus air) can strongly influence the measured relative abundance of emissions
Laboratory measurement of emissions	• Measurement of 204 trace gases and 30+ particulate matter components for southeastern and southwestern U.S. fuel types • Many of the 182 trace gases measured by GC–MS have been seldom measured in smoke previously
Field measurement of emissions	• Field measurements were well-correlated with laboratory measurements for EPA Criteria pollutants and many other trace gases supporting value of laboratory data • Potential firefighter fireline exposure exceeded peak levels but not per shift levels on control lines • FTIR technology can nonintrusively provide temporal measurement of trace gases at detection limits exceeding current standards

Table 2
Summary of airborne and ground-based sampling performed in the southeastern U.S. in February/March 2010 and November 2011. See [48, 49] for a more detailed description of the fires and the flight tracks associated with airborne measurements. Area in ha.

Fire name	Lat/Long (°)	Fuel type[a]	Area (ha)	Measures[b]
Camp Lejeune IA	34.5798 N, 77.3167 W	2yr	36	A, G
Camp Lejeune ME	34.6422 N, 77.4617 W	1yr, 2yr, cuh, uh	677	A, G
Little Florida 1	34.0708 N, 78.2780 W	wire grass, gallberry	16	A
Little Florida 2	34.0687 N, 78.2817 W	gallberry	24	A
Bear Pen	34.1287 N, 78.3388 W	grass		A
Holly Shelter	34.5467 N, 77.8367 W	lit	23	A
Fort Jackson 6	34.0247 N, 80.8711 W	longleaf	62	A, G
Fort Jackson 9b	34.0042 N, 80.8769 W	longleaf, sparkleberry	36	A, G
Fort Jackson 22b	34.0844 N, 80.7731 W	longleaf, loblolly, oak	29	A, G
Pine Plantation	33.5803 N, 81.1653 W	loblolly	16	A
Georgetown	33.2025 N, 79.4017 W	grass, uh	61	A
Francis Marion	33.2153 N, 79.4761 W	longleaf, wire grass	147	A
Bamberg	33.2347 N, 80.9447 W	longleaf, loblolly, uh	36	A

[a] Refer to [47] for explanation of fuel codes. Wire grass - *Aristida stricta*, gallberry - *Ilex coriacea*, longleaf – *Pinus palustris* litter, sparkleberry – *Vaccinium arboreum*, loblolly – *P. taeda* litter, oak – *Quercus* spp. leaves
[b] A – airborne, G – ground.

(9) a High Resolution Time of Flight Aerosol Mass Spectrometer (HR-TOF-AMS), (10) an Ultrafine Condensation Particle Counter (11) a Scanning Mobility Particle Sizer (SMPS), (12) a Fast Mobility Particle Sizer, (13) an Aerodynamic Particle Sizer, (14) a Micro-Orifice Uniform Deposit Impactor (MOUDI), and (15) a Mass Monitor (DMM) [40, 41, 45, 46]. Particulate matter was collected on glass and Teflon filters and subjected to a variety of offline measurements and analyses for elemental and organic carbon, elemental composition by X-ray fluorescence, ion chromatography analysis, levoglucosan and polycyclic aromatic hydrocarbons [41].

A total of 21 fuel beds representing six southeastern fuel types and 56 fuel beds representing eight southwestern U.S. fuel types were burned and measured using this suite of instruments [41, 47]. Three or four replications of fuel beds from Camp Lejeune (five fuel types) and five fuel beds of pine needles from Fort Benning were burned. Unburned residue was weighed at the end of each fire to determine consumption; however, some fires were extinguished after sampling was completed before all fuel was consumed in order to keep to the burning schedule.

2.3. Field measurement of emissions

The field component of the project consisted of deployments to Camp Lejeune (February/March 2010) and Fort Jackson (October/November 2011). Prescribed burns at the two locations ranged from 30 to 677 ha. Each burn contained one or more of the fuel types from the laboratory phase as well as additional types. The fuel types at Camp Lejeune represent fuels associated with frequent prescribed burning (every 1–3 years) which is the typical application in the coastal plain [1]. In response to managers' concerns, the fuel types at Fort Jackson were added to represent hazardous fuel accumulations resulting from no prescribed fire use on certain parcels.

Fort Jackson is in the Sandhills region, a band of ancient beach dunes separating the coastal plain from the Piedmont [26] (Fig. 1); the longleaf pine forests here are distinctly different from Camp Lejeune. Fort Jackson plots were specifically selected to include stands with significant understory growth, i.e. little or no recent prescribed burn history. One plot had not burned since 2003 and the other two plots had not been burned in at least 50 years. The older stand conditions at Fort Jackson, to some extent, serve as a living model of the fuel conditions expected if prescribed fire programs become further restricted across the southern United States. Burning during the relatively dry fall season was chosen to emulate weather conditions that might occur during higher intensity wildland fires to determine if there was an effect on emissions; however, fuel and weather conditions were such that this was not achieved. Our ability to estimate fire intensity from fuel consumption was severely limited. Ocular estimates of flame length are the best measure of fire intensity that we have for these burns.

Airborne measurements [48, 49] used a closed-cell FTIR instrument to collect and measure gases, a nephelometer measured light scattering which was correlated with particulate mass ($PM_{2.5}$) and other particulate sampling instruments. At some of the burns, whole air samples were also collected and analyzed [48]. At Camp Lejeune and Fort Jackson, a closed-cell Land based FTIR (the LaFTIR), based on the original unmodified spectrometer (MIDAC 2500) and detector (Graseby FTIR-M16) coupled with a smaller, vibration-isolated multipass Infrared Analysis, Inc. White cell, was used to estimate the composition of smoke emissions from residual smoldering combustion (RSC) of large woody fuels such as stumps and logs. RSC occurs frequently and in general refers to the smoke that is produced after the flame front has passed. This unlofted smoke is often associated with reduced highway visibility [20]. In addition to the LaFTIR closed cell instrument, at Fort Jackson a Bruker OPAG-22 OP-FTIR was set up at a fixed location for each burn, typically along a fuel break on the perimeter. The OP-FTIR sampled a mixture of both flaming and smoldering phase smoke as described below [42].

The comparison of emissions measured by various methods has been limited. In particular there are few papers which compare laboratory versus field measurements, and few that contrast airborne versus ground-based sampling [38, 48, 50, 51]. Ground-based emission measurements have been related to airborne measurements and modified combustion efficiency (MCE) has been used as the metric to correlate emissions from fires over several orders of size [52]. The design of the overall project allowed us to examine the relationship between emission factors derived from the laboratory phase with emission factors developed for similar fuel types in the field phase [51]. Ground-based smoke emissions and airborne emissions measured using the same type of instrument were compared. Using MCE to indicate fire phase, we compared agreement between laboratory and field-derived emission factors for similar fire phases (flaming, mixed, smoldering). The flaming phase is generally characterized by more complete oxidation and higher MCE values whereas the smoldering phase is characterized by less complete oxidation and lower MCE values. The data from the controlled laboratory burns were synthesized with the actual field measurements [51]. While we did

not include organic soils specifically collected in eastern North Carolina as a fuel type, organic soils from other locations were included in the laboratory phase and the results are potentially applicable to fires arising in the pocosin type which grows on organic soils [32]. Emissions from organic soil from eastern North Carolina and duff, partially decomposed forest litter material from longleaf pine [53], were compared with our ground-based LaFTIR results by converting emission ratios to emission factors using carbon mass balance [54, 55].

3. Results and discussion

To date, results from this project have been published in a variety of journals [40–42, 45–49, 51, 56–68] and most of the papers concentrate on either the laboratory or the field experiments. An archived database containing emission factors gleaned from five decades of literature as well as the emission factors derived from this work was also compiled and made available [69]. Here a brief description and discussion related to the fuel types in the project is presented prior to presentation of the results related to all three components of the project.

The fuels from Camp Lejeune used in the laboratory experiments consisted principally of five species of hardwood shrubs and small trees that grow in the southeastern Atlantic coastal plain: red maple (*Acer rubrum* L.),[1] redbay (*Persea borbonia* (L.) Spreng.), loblolly bay (*Gordonia lasianthus* (L.) Ellis), inkberry (*Ilex glabra* (Pursh) Chapm.), and fetterbush (*Lyonia lucida* (Lam.) K. Koch) [47]. Calculated bulk density of the various laboratory fuel beds differed by an order of magnitude between the five fuel types (Table 3). The loading for the 1-and 2-year rough fuel types fell within the range of published loadings for 1- and 2-year roughs under longleaf pine [16]. Loading of the pocosin fuel type, seldom sampled to determine fuel mass and structure [16, 32, 33], also fell within the range of reported loading for high pocosin and high brush fuel as did the understory hardwood fuel type (Table 3). Loading for the chipped understory hardwood (cuh) fuel type was similar to [17] depending on which 10 h fuel loading is used and within the range of the experimental fuel beds used by [70]. The understory hardwood (uh) fuel loading fell within the range of previously reported results; however, the bulk density of the laboratory fuel beds was much higher than the field fuel beds (Table 3) due to difference in depth between the reconstructed laboratory fuel beds and the naturally occurring fuel beds. The range of fuel loading in the litter fuel beds (lit) fell within the range of reported fuel loading for slash and loblolly pine [9].

Access to the field sites prior to the prescribed burns was severely limited due to ongoing military training activities. The February 2010 burn (IA) at Lejeune occurred in a two year rough of inkberry and fetterbush. The March 2010 burn (ME) included masticated fuels, resprouted fetterbush shrubs and understory hardwoods including red maple and sweetgum (*Liquidambar styraciflua* L.), an untreated moderate density understory of redbay, red maple, inkberry, and fetterbush, and an area of one-two year rough of fetterbush and swamp titi (*Cyrilla racemiflora* L.) with grasses. Understory fuels at Fort Jackson included farkleberry (*Vaccinium arboreum* Marshall) and longleaf pine needles. Limited fuel sampling occurred at Fort Jackson to estimate pre-burn mass per unit area and fuel mass consumed. With the exception of the IA burn at Camp Lejuene, sampling estimates of fuel consumption of the field burns were generally the same order of magnitude as the laboratory burns (Table 3). Further comparison of fuel consumption measurements was not possible.

3.1. Instrument, software and spectral database improvements

Inclusion of new hardware components of the FTIR spectrometer and gas cell installed at the FSL significantly improved detection capabilities for most chemical species [37]. This same spectrometer was subsequently reconfigured and deployed in the aircraft as a closed-cell system. The initial signal-to-noise ratio (SNR) reported for the original FTIR instrument was ~100 for the spectra examined [38]. In 2004 the SNR of the FTIR instrument was roughly 500, but this was increased to over 30,000 during the course of the project via improvements to the hardware, in particular interfacing a better infrared spectrometer. The increased sensitivity permitted FTIR detection of numerous new species in real-time such as HONO, glyoxal and glycolaldehyde [59, 61, 66].

The improved evaluation software [71] processed the data collected by the various FTIRs using a variety of analyses to isolate the individual spectra for chemical species. As a test of the changes to the improved software, the reference spectra developed by PNNL were evaluated. For eight of the nine chemical species used, the estimated variability in derived concentration was within 1–2% of the observed, which is less than the error associated with the IR intensity values as measured in the PNNL laboratory.

In terms of potential new vapor-phase species that could be analyzed via FTIR, the proposed chemicals identified using the four criteria described in Section 2 initially numbered ~50. The spectra of 53 chemicals were added to the PNNL spectral library (Table 4). We note that these spectra can be used with any IR spectrometer so, for example, they could potentially be used by space-based instruments e.g. [72] to measure smoke emissions in addition to the ground and aircraft-based platforms described here. All the infrared reference data derived from this project are now publicly available at nwir.pnl.gov. While many of these new gases have been detected in wildland smoke using techniques such as GC–MS and the spectra now exist, many of them fell below the detection limits of the improved FTIRs used in both the laboratory and field measurements (Table 1).

3.2. Laboratory measurement of smoke emissions

In some cases (pocosin (poc) and understory hardwood (uh) fuel types), the burn was terminated after the smoke had been sampled for a sufficient period of time (e.g. 20 min) to adequately characterize it. The fuel consumption that is reported does not reflect the consumption that might have occurred if the fire extinguished naturally. Note that the fuel moisture of these fuel beds fell in the range of dead fuels (unsaturated wood, $<35\%$); reported moisture content of live stems and foliage of these species ranged from 80% to 140% and from 100% to over 300% respectively [73].

The range of fuel loading in the litter fuel beds (lit) fell within the range of reported fuel loading for slash and loblolly pine [9] providing further support for the use of laboratory emission factors in field application. While we burned comparable fuel loadings in the laboratory and field experiments, differences in moisture content and bulk density affected the combustion efficiency of the different burns illustrating the challenges inherent in using complex wildland fuel beds in laboratory experiments e.g. [74, 75]. These differences are one of the primary reasons that laboratory and field experiments were compared and synthesized [51].

Other than the whole air canisters, most smoke measurements occurred in real-time and did not require offline chemical analysis. The canister anlysis method used gas chromatography (GC) coupled with either flame ionization detection, electron capture

[1] Source of binomial nomenclature is USDA, NRCS. 2014. The PLANTS Database (http://plants.usda.gov, 20 May 2014). National Plant Data Team, Greensboro, NC 27401-4901 USA.

Table 3
Characteristics of fuel beds burned in the laboratory (L) and in prescribed burns (F) to measure smoke emissions.

Fuel type[a]	Moisture content (%)		Preburn dry mass ($kg\ m^{-2}$)		Bulk density ($kg\ m^{-3}$)		Fuel consumption ($kg\ m^{-2}$)	
	L	F	L	F	L	F	L	F
1yr	21.7	nm[b]	0.67		0.67		0.62	
2yr	27.4	nm	0.72		0.72		0.54	
cuh	5.6	nm	2.67		2.67		2.54	
uh	12.5	nm	2.02		2.02		1.19	
poc	12.1	nm	4.63		4.63		2.50	
lit	5.0	nm	1.18		8.47		0.89	
IA				nm		0.85		0.09
ME				nm		1.68		1.02
Block 6				nm		nm		0.86
Block 9b				nm		0.85		0.57
Block 22b				nm		1.12		0.47

Pre-burn loading not measured, value presented is actual estimated fuel consumption.

[a] 1yr=1 year rough (57% dead material), 2yr=2 year rough (73%), cuh=chipped understory hardwood (96%), uh=understory hardwood (50%), poc=pocosin (27%), lit=pine litter (100%). Field plots were a mixture of fuel types. Plot IA=2yr, ME=1yr, 2yr, cuh, uh, Block 6=54 year old longleaf pine fuels, Block 9b=55 year old longleaf fuels, Block 22b=8 year old mixed longleaf, loblolly pine, and oak litter. See [47, 48] for a detailed description of the species composition of fuel type.

[b] nm=not measured.

Table 4
Chemical species found in wildland fire smoke added to the PNNL spectral library [57].

1-Pentanal	3-Methylfuran	Hexadecane	Methyl-2-methylbutyrate
1-Penten-3-ol	3-Pentanol	Hexyl acetate	Myrcene
2,3-Butanedione	4-Penten-1-ol	Hydrogen peroxide	Naphthalene
2,3-Dimethylfuran	5-Nonanol	Isobutyric acid	Octanoic acid
2,5-Dimethylfuran	Acetol	Isocaproic acid	Pentadecane
2-Carene	Acrylamide	Isopentanal	Phenol
2-Methoxyphenol	Acrylic acid	Isopentyl acetate	α-Pinene
2-Methylfuran	Diacetone alcohol	Limonene	β-Pinene
2-Nonanone	Ethyl benzoate	Malonic acid	Propylene carbonate
2-Pentanone	Eucalyptol	Menthol	Syringaldehyde
2-Pentyl furan	Geraniol	Methyl acetoacetate	Valeric acid
2-Vinyl pyridine	Glycolaldehyde	Methyl glyoxal	Vinyl phenol
3-Carene	Glyoxal	Methyl vinyl ether	Vinyl toluene
3-Methoxyphenol			

detection, or quadrupole mass spectrometer detection. The canister methods were important so as not to miss those chemicals liberated as the reactions occurring in the flame and smoke plume sometimes continued for hours and days after release e.g. [76].

Combustion efficiency of most experimental burns was relatively high (as characterized [77] by MCE of 0.93–0.96) indicating primarily flaming combustion and the resulting associated compounds. The pine litter fuel beds tended to smolder resulting in a lower MCE of 0.89 and a greater production of compounds associated with incomplete combustion such as methanol, acetic acid and furan [49]. We note that nitrous acid (HONO), an important precursor to OH, was observed for all fuel types [45]. In the FSL burns, oxygenated volatile organic compounds (OVOCs) accounted for an average of 66% of the total non-methane hydrocarbons (NMHC) measured in the Camp Lejeune fuels. Trace gas emission factors from the laboratory experiments in 2009 were generally lower than emission factors measured in the field [55] with an airborne FTIR instrument at Camp Lejeune in 1997; MCE of the previous prescribed burns was lower than the lab experiments [55]. The OP-FTIR used in the lab measured 10 additional trace gases in comparison with the earlier work, largely due to improved sensitivity.

The carbonaceous particulate matter produced by combustion can be separated into two categories – elemental carbon (EC) and organic carbon (OC) even though the definitions of elemental carbon, black carbon, brown carbon, etc. are not always rigid [78]. These two emissions (EC and OC) comprise total carbon. With the exception of the chipped understory hardwoods, the emission factors for OC were higher in the southeastern fuels when compared to southwestern fuels [41] even though the MCEs were similar. The litter fuels produced the greatest emission factors for OC consistent with an observed inverse correlation between MCE and OC. Smoldering combustion tends to produce more products of incomplete combustion (OC) and more fine particles ($PM_{2.5}$) [79, 80]. The laboratory results supported this observation. The correlation between EC and combustion efficiency was very weak; however, the ratio of EC to total carbon was found to increase markedly when the MCE exceeded 0.94. In total, 204 trace gases, many of which had seldom been reported in wildland fire smoke, and 30+ components of particulate matter were measured in the laboratory phase of this project (Table 1).

3.3. Field campaigns

As the late fall and winter is the active time for prescribed burning in the southern U.S., several airborne measurements were taken from prescribed burn smoke plumes in the vicinity of each installation [42, 48, 49]. The prescribed burns at the installations contained a mixture of the fuel types which had been measured in the 2009 laboratory experiment as well as additional fuel types. Firing patterns at both Camp Lejeune and Fort Jackson were selected to increase the fire's energy release rate. Helicopter ignition using the Delayed Aerial Ignition Device (ping-pong ball) system [81] increased the energy release rate on the Camp Lejeune burn to compensate for the short burning window that day and the wet conditions of the understory vegetation. The burn had to be completed by midafternoon to minimize smoke impacts on an adjacent major highway. Even though the energy release rate was increased, the overall consumption in the IA burn was only 11% (0.09/0.85) (Table 3). The March burn (ME) was hand-ignited with drip torches and included several fuel types. Wide strips were used on the edge of the unit to generate sufficient flame and intensity to carry the fire through wetter and denser fuel conditions in the interior of the burn unit. The fuel consumption in the ME burn was much higher (61%). We observed no difference in the

airborne emissions from the beginning of the burn (initially dominated by the masticated fuels) and the transition into the untreated heavier fuels in the interior of the unit. The fire-averaged MCE of 0.94–0.95, similar to the laboratory burns, indicated that relatively efficient burning and the desired intensity were achieved.

Hand-ignition with drip torches was used to start the burns at Fort Jackson. As the intent was to generate fires with greater intensity (energy release rates) than at Camp Lejuene, spacing between adjacent strip head fires was adjusted throughout each day as ambient conditions changed to increase intensity while minimizing damage to the overstory pines. Due to a lack of fire behavior observations, estimated burning conditions (Table 5) were based on calculated indices from the 1988 National Fire Danger Rating System [82], a version specifically modified for use in the southern U.S. [83]. The indices are general indicators and should not be assumed to represent actual fire behavior of individual fires or prescribed burns wherein firing patterns are used to control fire behavior [1, 84]. Observed weather data at Fort Jackson and Camp Lejeune were used to calculate fuel moisture content of 1 h, 10 h, 100 h, and 1000 h timelag fuels and fire behavior indices were then calculated for fuel model D "Southern Rough". We assumed that fuel model D reflected the fuel accumulation in the burn units at Fort Jackson resulting from long term fire exclusion. Because neither base had a weather station with a sufficiently long record to determine the climatology (cumulative distribution function) of the fire danger indices [85], the calculated indices were compared to the climatology of nearby locations – Carolina Sandhills National Wildlife Refuge for Fort Jackson and Sandy Run remote automated weather station for Camp Lejeune. The percentile values of the indices are contained in Table 5.

Comparison of the data from the Fort Jackson and Camp Lejeune burns showed the air temperatures were higher at the time of burning at Fort Jackson while wind speeds were comparable. The NFDRS calculations show lower fire danger indices at the time of the Fort Jackson burn but also show lower dead fuel moisture content. The apparent inconsistency between the fire danger indices and dead fuel moisture can be attributed to live woody fuels at the time of year when the burns were conducted. The burns at Camp Lejeune were conducted in February and March. At this time of year live woody moisture content is normally at its lowest and these fuels would be expected to actively support burning [86]. In contrast, the burns at Fort Jackson were conducted in late October and early November. At this time of year near the end of the growing season the live woody fuel moistures are normally still high and the live fuels would moderate fire behavior even though the Keetch–Byram Drought Index (KBDI) indicated some rainfall deficit. We cannot conclude that the burns at Fort Jackson were "higher intensity" burns in comparison to Camp Lejeune. Instead, we assumed that the fires were similar in behavior and are representative of the range of prescribed fire. Anecdotal observations of flame length at Fort Jackson indicated that heading flame lengths were generally less than 1 m in the litter dominated areas and up to 3 m in areas with dense shrubs [87]. Backing fire flame lengths were generally less than 0.5 m. Longer flame lengths and higher heat release occurred in areas where a backing fire merged with the adjacent strip head fire or where two flanking fires merged [81, 88].

Table 5
Estimated fuel moisture content and fire behavior indices for prescribed burns.

Variable[a]	Location[b]					
Date	FJ 10/27/11	FJ 11/1/11	CS Percentile	CL 2/10/10	CL 2/11/10	SR Percentile
Air Temp. (°C)	26	18	61, 34	8	13	7, 16
Wind (kph)	14.8	9.8	86, 55	14.8	8.2	90, 51
1 h (%)	5.4	4.8	53, 41	5.9	4.9	15, 6
10 h	7.0	6.4	35, 16	8.1	6.8	21, 3
100 h	13.4	12.9	20, 12	16.9	14.1	23, 5
1000 h	20.1	19.5	65, 55	24.7	20	73. 30
IC	18	19	66, 68	30	28	93, 92
SC	7	7	50	30	18	95, 79
BI	31	31	49	85	69	95, 85
KBDI	184	208	44, 48	2	14	2, 6
ERC	21	24	48, 54	48	51	91, 96

[a] Moisture content (in percent for 1, 10, 100 , 1000 h timelag classes) and NFDRS indices estimated using 1988 National Fire Danger Rating System [82]. IC = ignition component, SC = spread component, BI = burning index (÷ 32.8 = flame length (m)), KBDI = Keetch–Byram Drought Index, ERC = energy release component.

[b] FJ = Fort Jackson, CL = Camp Lejeune, CS = Carolina Sandhills National Wildlife Refuge, SR = Sandy Run remote automated weather station. [85].

The airborne MCE for Fort Jackson burns was on average slightly lower (0.92–0.94) but still within the range observed in the lab. Excluding the Pine Plantation burn, the airborne MCEs ranged from 0.93 to 0.96 for the other fires. Due to the location of the ground-based FTIR instruments (along the fireline within close proximity to the backing fire used to secure the fireline or adjacent to smoldering stumps), the ground-based MCE was appreciably lower (0.79–0.88) in the Fort Jackson burns for the two ground-based FTIR systems.

A total of 97 trace gas species were quantified from both airborne and ground-based sampling platforms at Fort Jackson using the FTIR instruments and offline analysis of whole air samples (WAS). The known chemistry of the monoterpenes and their measured abundance of 0.4–27.9% of non-methane organic compounds (NMOCs) and ~21% of organic aerosol (mass basis) in the sampled plumes suggests that the terpenes impacted both secondary formation of ozone (O_3) and aerosols, as well as small organic trace gases such as methanol and formaldehyde in the first few hours after emission. The variability in the initial terpene emissions in the Fort Jackson fire plumes was high and, in general, the speciation of the initially emitted gas-phase NMOCs was 13–195% different from that observed during the Camp Lejeune deployment. This variability is similar to that observed in other studies in conifer forests [48]. It is likely that differences in stand structure and environmental conditions contributed to the high variability observed within and between these studies. While isoprene and terpenes are all alkenes, hardwood trees emit isoprene almost exclusively whereas pine trees naturally emit primarily terpenes (such as α- and β-pinene, limonene) e.g. [89].

We did not quantify the overstory species composition at Camp Lejeune and Fort Jackson, but the dominant overstory species were pines with a mixture of hardwoods. As expected, the $\Delta HCN/\Delta CO$ emission ratio was found to be fairly consistent with previous airborne fire measurements in other conifer-dominated ecosystems. It was possible to measure the downwind chemical evolution of the plume from one Fort Jackson burn and three prescribed burns in South Carolina that were only sampled with the aircraft. Significant O_3 formation occurred in all of these plumes within two hours. Changes in plume composition are also important to consider as the smoke is transported outside of the immediate vicinity of a prescribed burn and can have unintended impacts on regional air quality and human populations some distance from the fire location [90, 91].

Three different FTIR instruments were deployed at Fort Jackson (two closed cell: the airborne and the roving LaFTIR, and one open-path system, the OP-FTIR along the fireline). Comparison of these measurements confirmed earlier results and led to some new key findings: the FTIR data show that the method (airborne, ground) used to sample smoke can strongly influence the relative abundance of the emissions that are measured [50]. For these fires, the majority of fire emissions were lofted sufficiently high enough in the convection column so that they could be sampled by the

airborne FTIR – which had a minimum distance above ground level of 500 ft (152 m). The mobile, ground-based, point sampling FTIR measured the contribution of individual smoldering stumps and trees scattered throughout the burn site, and these data were characterized by significantly lower MCE values (associated with RSC). The OP-FTIR, however, provided a ~30 m path-integrated sample of emissions transported across the fireline and typically measured two distinct combustion regimes: (i) a flaming phase immediately after the adjacent line ignition (back firing to establish burnt buffer) but before the adjacent plume achieved significant vertical development, and (ii) a smoldering phase which consisted of primarily RSC smoke wafting across the fireline. The OP-FTIR provided our best estimate of fireline exposure to smoke for wildland fire personnel who are charged with holding the control lines. Our data suggest [46] that peak exposures to smoke are more likely to challenge permissible exposure limits for wildland fire personnel than shift-average (8 h) exposures which supports earlier work wherein firefighters were equipped with monitoring devices to determine smoke exposure during prescribed and wildland fires [92]. In particular, avoidance of areas with high levels of residual smoldering combustion or airway protection by fire personnel may be possible methods to mitigate exposure to peak levels [42].

As mentioned above, the project further allowed us to compare emission factors determined in a laboratory setting with emission factors determined in the field [51]. In comparison with the laboratory, a broad variety, but a smaller number of chemicals (21 trace gas and $PM_{2.5}$) was measured in the field. Due to the sensitivity of the OP-FTIR instrument and software improvements, the laboratory emission factors originally reported [47] were recomputed using carbon mass balance which resulted in slight changes [51]. The revised trace gas emission factors can be found in Table 6 and the full set of recomputed emission factors are supplemental material available at http://www.atmos-chem-phys.net/13/89/2013/acp-13-89-2013-supplement.zip.

For most of the chemicals NO_x to $PM_{2.5}$ the mean lab EF/field EF ratio for the pine understory fuels was not significantly different from unity [51]. Based on the lab/field comparison, emission factors for 357 pyrogenic species (including unidentified species) for southern pine understory fuels and organic soils were developed and recommended as useful for field application. The emissions we measured using the LaFTIR instrument with focused sampling on a variety of individual fuel types prone to produce residual smoldering generally fell within the ranges previously measured for bog soils and longleaf duff (Table 7).

While the number of chemical species detected in the laboratory phase with the large suite of instruments that was deployed far exceeded the number of species measured in the field phase, the FTIR instruments used were able to non-intrusively sample an increased number of species at lower concentrations than were available only 4 years ago. The FTIR technology also provided continuous measurement in comparison to collected gas canisters which generally lack a continuous temporal signature and may change chemically from the time of collection to the time that analysis is performed. Many new chemical species previously undetected by FTIR were observed, some with good signal-to-noise, others near the detection limits of the instruments. Not all were reported in the literature, but include such species as HONO, phenol, glyoxal, methylglyoxal, glycolaldehyde, pinenes, limonene, etc. FTIR technology has the potential to improve real-time measurement of trace gas emissions in field settings (Table 1).

3.4. Emission factors database

Perception of a lack of information on emissions from southern fuel types resulted in a search through the published and "grey" literature to locate emissions information. The oldest reports date back to the 1960s, which first reported CO and CO_2 emission factors. It was not until the early 1980s, however, that significant amounts of other emissions data for wildland fire were reported. In total, 230 references were located and evaluated. See reference [69] for a complete listing of the documents which were located and included in the database. Emissions information for 118 smoke components from 51 of the reviewed references are currently contained in Wildland Fire Emissions Factor Database which contains information published up to 2011 [69]. Future updates to the database may occur.

Pertinent information related to the fuels, the instrumentation used, and geographic location is all contained in the spreadsheet. All 230 references were electronically scanned and are available. If it was not possible to convert emission ratios and emission concentrations to emission factors using information contained in the

Table 6
Relationship between laboratory and field derived emission factors for southern pine forest understories. The lab average was calculated from all of the southeastern fuel beds as described in [47] with carbon mass balance calculations resulting in slight changes [51] since more carbon was accounted for; field average was based on the fires in Table 2. Predicted lab EFs used mean field MCE of 0.936. See [51] for a complete description of the analysis approach used to derive the lab/field EF ratios.

Species	Lab_{meas} (g kg^{-1})	$Field_{meas}$ (g kg^{-1})	Lab_{pred} (g kg^{-1})	Lab_{meas} /$Field_{meas}$	Lab_{pred} /$Field_{meas}$
CO_2	1738	1668		1.08	
CO	83.08	72.1		1.15	
MCE	0.932	0.936		1.00	
NO	1.78	0.88		2.02	
NO_2	1.01	2.68		0.38	
NO_x as NO	2.44	2.55	2.42	0.96	0.95
CH_4	2.79	3.02	2.61	0.93	0.86
C_2H_2	0.18	0.30	0.18	0.61	0.59
C_2H_4	0.90	1.16	0.85	0.77	0.73
C_3H_6	0.35	0.40	0.32	0.86	0.80
HCHO	1.11	1.51	1.02	0.74	0.67
CH_3OH	1.02	1.05	0.93	0.97	0.88
HCOOH	0.25	0.09	0.23	2.65	2.40
CH_3COOH	2.20	1.32	2.03	1.67	1.54
C_6H_5OH		0.33			
C_4H_4O	0.20	0.20	0.17	0.99	0.87
$C_2H_4O_2$		0.25			
HCN	0.29	0.59	0.26	0.49	0.43
NH_3	0.75	0.50	0.72	1.50	1.43
HONO	0.38	0.52	0.38	0.73	0.73
$PM_{2.5}$	6.63	13.55	6.88	0.49	0.51

Table 7
Emission ratios (C_i/C_T) converted to emission factors (EF, g kg^{-1}) for southeastern fuels not measured during the laboratory phase of this study. See [53] for a complete description. LaFTIR samples included burning stumps, dead/down debris, fatwood, and duff [48].

	NC Bog soil		Longleaf duff		LaFTIR
Species	C_i/C_T	EF	C_i/C_T	EF	EF Range
CO_2	72.2	1693	65.1	1207	1305–1554
CO	23.9	356.6	26.4	311.4	140–222
CH_4	1.5	12.79	1.85	12.471	5.20–11.50
C_2H_4	0.179	2.67	0.192	2.265	0.89–1.53
C_2H_2	0.012	0.17	0.026	0.285	0.22–0.25
C_3H_6	0.155	3.47	0.17	3.008	
HCHO	0.073	1.17	0.784	9.910	1.79–2.51
Glycolaldehyde	0.014	0.45	0.183	4.626	
Formic acid	0.063	1.54	0.316	6.125	
Acetic acid	0.231	7.39	0.855	21.614	1.03–3.84
CH_3OH	0.226	3.85	1.095	14.764	2.35–6.42
C_6H_6O	0.068	3.41	0.219	8.674	

original source, the data were not included in the database. Some of the references include sufficient information so that experimental error (standard deviation) could also be determined. This information was not included in the spreadsheet version due to space limitations. Inclusion or exclusion of data from a particular report is not a statement on the quality of the information. Users of the database will need to draw their own conclusions regarding the applicability of the information to their particular situation.

4. Summary

The SERDP-sponsored project enabled new technologies and furthered basic science, mostly in the area of infrared spectroscopy, a nonintrusive broadband (remote sensing, depending on configuration) measurement method well suited to characterize smoke from wildland fire. Advances in hardware, software and supporting reference data realized a nearly 20 × improvement in sensitivity and now provide quantitative IR spectra for potential detection of 53 new species and actual field quantification of several new species such as nitrous acid, glycolaldehyde, α-/β-pinene and D-limonene. The new reference data also contribute to the determination of the global warming potential of greenhouse gases by both (1) enabling detection of their ambient concentrations, and (2) quantifying their ability to absorb IR radiation, i.e. act as a greenhouse gas.

Another key deliverable resulting from our studies are the data consisting of hundreds of the best possible measurements of EF for a very broad suite of both trace gases and particulate species for southeastern U.S. fuels. The list of chemical species measured in this project is the most extensive smoke characterization achieved to date and is presented in extensive tables of EF in the various publications [40–42, 45–49, 51, 56–68], including summaries by vegetation type, e.g. semiarid shrublands or pine understory, since the project was one of three related efforts. Prescribed fire managers can use the emission factors and fuel loading data compiled in this project when planning prescribed burns. Uncertainty in the emissions information can be coupled with uncertainty in fuel loading and consumption derived from other sources to estimate the uncertainty of emissions produced [93]. Emission ratios (ER) of various compounds to CO were derived and can be used by air quality specialists to estimate downwind concentrations and photochemical changes for certain species using only data from CO monitors. Multiple measurements of in-plume chemical transformations, O_3 formation, and secondary organic aerosol formation can be used by modelers to refine air quality models such as CMAQ [96] to improve estimates.

While field studies results are the most desirable, we have also confirmed that studying laboratory biomass fires can significantly increase our understanding of wildland fires, especially when laboratory and field results are carefully combined and compared. In particular, we have found that many of the emission factors for the unlofted emissions that are produced by residual smoldering combustion are very similar between the laboratory and field measurements, thus making laboratory studies of great value at characterizing smoldering-phase emissions. These unlofted emissions are of primary concern to prescribed fire managers because of the potential for traffic corridor obscuration and effects on human health. Characterization of such emissions will enable us to improve current models predicting low visibility and super-fog formation [20, 94].

In summary, this five year study provided a wealth of data and technology which can be used to improve our use of prescribed burning, better manage carbon resources, and improve prediction of both the local and global effects of fire in fuels of this or similar types.

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