Air emissions from organic soil burning on the coastal plain of North Carolina

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HIGHLIGHTS

► $PM_{2.5}$ EFs were at least a factor of 2 greater than those from above-ground fuels.
► CO EFs (250–300 g kg^{-1} fuel dry weight) are at the high end of the range of previously published EFs.
► Levoglucosan was found to compose 1–3 percent of $PM_{2.5}$ from the organic soil fires.
► $PM_{2.5}$ emissions may account for 10–20% of the total U.S. $PM_{2.5}$ air emission inventory.

ARTICLE INFO

ABSTRACT

Emissions of trace gases and particles ≤2.5 microns aerodynamic diameter ($PM_{2.5}$) from fires during 2008–2011 on the North Carolina coastal plain were collected and analyzed. Carbon mass balance techniques were used to quantify emission factors (EFs). $PM_{2.5}$ EFs were at least a factor of 2 greater than those from forest burning of above-ground fuels because of extended smoldering combustion of organic soil layers and peat fuels. This is consistent with CO_2 EFs at the low end of previously reported ranges for biomass fuels, indicating less efficient combustion and enhanced emissions of products of incomplete combustion (PICs). CO EFs are at the high end of the range of previously published EFs for smoldering fuels. The biomass burning tracer levoglucosan was found to compose 1–3 percent of $PM_{2.5}$ from the organic soil fires, similar to fractions measured in smoke from above-ground fine fuels reported in previous studies. Organic soil fuel loads and consumption are very difficult to estimate, but are potentially as high as thousands of tonnes ha^{-1}. Combined with higher emission factors, this can result in emission fluxes hundreds of times higher than from prescribed fires in above-ground fuels in the southeastern US. Organic soil fuel represents a source of particles and gases that is difficult to control and can persist for days to months, jeopardizing human health and incurring considerable costs to monitor and manage. Extended fires in organic soils can contribute substantially to $PM_{2.5}$ on CO emission inventories and may not be adequately accounted for in current estimates.

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1. Introduction

The United States Environmental Protection Agency (EPA) has recently revised the National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$. Primary $PM_{2.5}$ standards are set at 15 μg m^{-3}, annual mean, and 35 μg m^{-3}, 24-h average, to provide increased protection against a wide range of PM-related health effects. These health effects include premature mortality, increased hospital admissions and emergency room visits, increased respiratory symptoms and disease, decreased lung function, and alterations in lung tissue and structure and in respiratory tract defense mechanisms (Fowler, 2003). Through emission of $PM_{2.5}$ and ozone

precursor gases, fire also reduces visibility. Hence, natural area and agricultural land management, nationwide, may come under increased scrutiny as regulators seek reductions in pollutant emissions which contribute to NAAQS violations. Current guidelines allow flexibility in application of prescribed burning in smoke-sensitive areas if burn prescriptions are adhered to. Biomass burning in North America can also be a potentially significant source of radiatively active trace gases (Vose et al., 1997; Wiedinmyer et al., 2006). Areas burned in North and Central America can exceed 10 million hectares per year, resulting in trace gas and PM emissions that range from 10 to 40% of total emissions from all sources (Hoezelmann et al., 2004; Wiedinmyer et al., 2006).

Residual smoldering combustion (RSC) has been found to produce elevated $PM_{2.5}$ (McMahon et al., 1980) and (non-CO_2) trace gas emissions relative to flaming combustion (Bertschi et al., 2003). The largest potential pool of carbon vulnerable to RSC in many areas

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is organic soils (McMahon et al., 1980; See et al., 2007). On the coastal plain of the eastern U.S., wildland fires that burn into organic soils can smolder for weeks or even months, emitting tons of air pollutants and posing health, safety, and ecological hazards to local communities. Although above-ground fuel loads and emission factors have received considerable attention in the laboratory and field, much less data are available for below-ground fuel components. Here we investigate CO_2, CO, and $PM_{2.5}$ emission factors (EFs) from smoldering organic soils in North Carolina, USA.

2. Site description and burn conditions

2.1. Pocosin Lakes National Wildlife Refuge (PLNWR)

The tracts burned at this site (see Plate 1) were occupied primarily by pond pine (*Pinus serotina*) pocosins with a heavy understory of loblolly-bay (*Gordonia lasianthus* L.), red bay (*Persea borbonia* L.), gallberry holly (*Ilex coriacea* (Persh) Chapm.), fetterbush (*Lyonia lucida* Lam. K. Koch) and wax myrtle (*Myrica cerifera* L.). A prescribed fire was ignited at 12 PM on February 16, 2008 with fine fuel moisture ranging between 9 and 10% and relative humidity at 50%. Fuels burned vigorously with flame lengths often exceeding 15 m. Pre-fire fuel loads in the pocosin ecosystem were highly variable. In the fetterbush/gallberry shrub patches, shrub density in

these species was quite high prior to burning ($\sim$9 tons acre^{-1} or 20 tonnes ha^{-1}, Mickler et al., submitted for publication). Pine and broadleaf litter and shrub comprised about 90% of the fuels consumed (11.2 tons acre^{-1} or 25 tonnes ha^{-1}), consistent with North Carolina Division of Forest Resources (North Carolina Smoke Management Program, 2008), which estimates fuel loads of 11–22 tonnes ha^{-1} for medium density tall brush (>1.3 m) composed of red bay, loblolly-bay, gallberry, and wax myrtle. Fuel consumption efficiency was high at 70 and 80% (6.2 tons acre^{-1}, or 14 tonnes ha^{-1}) of the shrub and litter fuel, respectively. Approximately 10% of the total fuels consumed were woody. Fuel loads and consumption in wax myrtle woodlands at PLNWR were lower than in the Fetterbush/gallberry stands. Only 1.2 tons acre^{-1} (2.7 tonnes ha^{-1}) was consumed in wax myrtle litter and woody fuel classes (Mickler et al., submitted for publication).

Following the prescribed burn, small pockets of smoldering combustion as deep as 1 m into the organic peat layers was observed. We collected smoke samples from these sites on February 17, 2008, and also removed over 60 kg of organic soil (peat) layers intact in cores 30 cm in diameter and 40–50 cm in length. These cores were collected fully intact and within a meter of areas where soil had burned to a depth of 50–100 cm. The cores were used to determine organic soil and elemental composition and to perform laboratory combustion tests.

Plate 1. Image of eastern North Carolina showing fire locations for this study. Inset shows burn scars from 2008 PLNWR and 2011 ARNWR wildfires within white rectangles. Imagery courtesy of Google Earth.

On June 1, 2008, a lightning strike in the PLNWR ignited the Evans Road Wildfire during a record drought season. This fire eventually consumed fuels over a 16,808 ha area, and soils smoldered for nearly three months. These ground fires were scattered randomly over the refuge, but where they occurred, they burned as deep as a meter or more. On August 12, 2008, smoke samples were collected from a smoldering peat seam on the PLNWR.

2.2. Green Swamp Preserve

The Green Swamp Preserve consists of 36,300 ha in southeastern NC (Brunswick and Columbus Counties). The area consists of pine pocosins and longleaf pine (*Pinus palustris*) stands on sandy peat soils. Following prescribed burning on Feb 10, 2009, small pockets of soil combustion were observed on Feb. 11. We collected smoke samples from these smoldering soil sites, which were usually less than a few m^2 in area and less than 50 cm deep. We also sampled smoke during the prescribed fires later that afternoon in pocosin habitat.

2.3. The Alligator River National Wildlife Refuge (ARNWR)

The Alligator River National Wildlife Refuge (ARNWR) and Dare County Air Force and Navy Bombing Range (DCBR) comprise 61,600 and 18,860 ha, respectively on the coastal plain of North Carolina (see Plate 1). These units have no water management capability, so water levels are dependent upon precipitation and tidal dynamics in Pamlico Sound. The primary vegetation type (83–100% of canopy cover) on the ARNWR is pond pine (*P. serotina*) with a shrub understory. Soils are histisols comprised of 2–3 m of organic muck which overlays marine sandy sediments. Pond pine canopy closure averages 70%, and the understory/midstory is comprised of wax myrtle, fetterbush, red bay, gallberry holly, and greenbrier (*Smilax rotundifolia* L.) with density ranging from moderately open to very dense. Scattered red maple (*Acer rubrum*), sweetgum (*Liquidambar styraciflua* L.), red and loblolly-bay, and black gum (*Nyssa sylvatica* L.) are present. A pine needle layer 2–15 cm deep covers the ground surface with some areas of heavy needle drape and ladder fuels, creating conditions which allow surface fire to spread to the overstory pine canopy. NCDFR (2008) gives a range in fuel loads of 4–8 tons acre^{-1} (9–18 tonnes ha^{-1}) for these pine/hardwood litter fuels.

Wildfire was ignited by lightning within the ARNWR on May 5, 2011. This fire eventually covered over 18,330 ha, and ground fires continued to smolder well into July. Forest Service officials reported that ground fire burned to a depth of ~45 cm, exposing root systems of trees. Our post-fire transects indicate that soils were burned to a depth 15–30 cm, with pockets of deeper soil consumption near the base of trees. Ground fire covered approximately half of the burned area. On May 12, 2011, air samples were collected near Highway 264, approximately 2 km south of Stumpy Point, NC. Flames were not visible, but smoldering soil combustion and smoke were widespread over the ARNWR. Samples were collected from 1 to 3 PM in smoke approximately 50–150 m from the combustion zones, and then in the late afternoon (3:30–5:30 PM) a separate set of samples was collected within 10 m of smoldering soil combustion.

3. Measurement methods

3.1. Portable field system

A backpack-portable sampling system which uses twin polished aluminum PM$_{2.5}$ cut personal mini-cyclones attached to carbonate plastic filter holders served as the inlets. Quartz fiber (for analysis of total gravimetric mass and organic PM$_{2.5}$ components) or Teflon filters (for total gravimetric mass, inorganic ions and elements) were used for PM$_{2.5}$ sampling. Filters were 37 or 47 mm diameter, and the quartz fiber filters (Pall Gelman, Ann Arbor, MI, USA) were baked at 550 °C for 24 h to remove organic contaminants prior to sampling. A 3-SLPM LICOR (Lincoln, NE, USA) pump pulls air through one of the cyclone/filter inlets, through 6 mm Bevaline, and then pushes through a flow restrictor (0.8 SLPM) to a LICOR 840 CO$_2$/H$_2$O infrared gas analyzer. The LICOR CO$_2$ and H$_2$O instrument showed linear response and was consistently within ±2% (usually 1%) of target calibration values over the range of 300–3000 ppm for CO$_2$ and 0–70 parts per thousand for H$_2$O vapor. A 5-SLPM diaphragm pump (BRC Rasmussen, Portland, OR, USA) pulls through the second cyclone/filter, and then pushes through a "T". On one side of the "T" a Photovac portable FID system, which has its own internal pump, samples at 1 SLPM. The FID was used in both total VOC mode, and also in a CH$_4$ only mode by using an activated charcoal filter immediately upstream of the FID unit. The CH$_4$ and total VOC measurements are the most uncertain of the trace gas measurements since they are performed by integrated canister sampling or via the portable FID, which has variable response factors for many of the compounds typically found in biomass burning plumes. Neglecting organic compounds would be expected to have a small (<5%) impact on CO$_2$, CO, and PM EFs because less than 5% of carbon mass from fires is emitted as CH$_4$ and VOC (Akagi et al., 2010; Andreae and Merlet, 2001). When the FID was not deployed, TENAX cartridges were used to sample air at 45 mL min^{-1} using an SKC (Eighty Four, PA, USA) Pocket Pump. On the other side of the "T", air is pushed through a restrictor to an H$_2$SO$_4$ reaction cell for CO detection (deployed at a flow rate of 45 SmLPM), calibrated against a compressed CO standard and also compared in laboratory studies to a gas-filter correlation system described below. The H$_2$SO$_4$ reaction cell produces an electric current as CO in air drawn over the H$_2$SO$_4$ bed is oxidized at one electrode to carbon dioxide while oxygen is consumed at the other electrode. Measurement of the current gives a measure of the concentration of CO in the air sample. The H$_2$SO$_4$ reaction cell is reasonably accurate (±10% or better) and produces a linear response to CO concentration. It can be operated by small (AA) batteries and has a lifetime of 1–5 years depending on exposure regime. The technique compared well for CO concentrations observed in smoke (within four to ten percent, with a slight negative bias) versus the gas filter correlation method (GFC) employed by a Thermo Electron Model 48C GFC Ambient CO analyzer (Thermo Environmental Instruments, Inc., Franklin, MA). At ambient levels it is more variable, within 30% of GFC CO values from 200 to 1000 ppb.

A temperature probe was positioned between the two PM inlets and logged to a Hobo (Bourne, MA, USA) data logger, which also logged ambient temperature. CO$_2$ and CO data were also logged at two second intervals to the Hobo data logger. Pumps and flow control devices used for sampling were calibrated with a DryCal (Butler, NJ, USA) Flowmeter, which is a primary calibration standard. All filters were conditioned for 24 h at 25 °C and relative humidity of 38% prior to tare weight determination (microbalance model MC5; Sartorius Corp., IL, USA). Following sample collection, filters were again conditioned for 24 h at 25 °C and relative humidity of 38% prior to final weight determination. PM$_{2.5}$ concentration was calculated by subtracting the tare weight from the final weight and dividing by the air sample volume. Filter blanks were also weighed before and after measurements at each site. Blank weight gains were less than 0.1% of sample PM$_{2.5}$ mass and were not significantly different from zero. Organic carbon to total carbon (OC/TC) ratios were determined from the PM$_{2.5}$ samples on quartz fiber filters using a Sunset (Hillsborough, NC, USA) analyzer as described in Hays et al. (2002) and Geron (2009).

Sampling inlets were located on the perimeter downwind of the burns areas far enough from combustion zones (5–100 of meters) to allow smoke plumes to cool to approximately ambient temperatures to allow partitioning of semivolatiles between gas and aerosol phase. Some smoke from prescribed fires in above ground fuels was also collected and is discussed here for comparison, but details are in a separate paper (Geron et al., in press). EFs from above-ground fine fuels are consistent with values published for prescribed fires in Vose et al. (1997).

3.2. Prescribed fire and laboratory methods

The burn hut laboratory used to test peat and other fuels is described in Gullett et al. (2008) and Hays et al. (2002) and other references cited therein. During our laboratory peat tests, the soil sample core integrity was maintained, so the fuel density and array appeared to represent that observed in the field. The peat cores were placed on a pan containing sand, which was in turn placed on a balance which allowed us to continuously monitor fuel loss. Ignition was performed using three methods which included a glowing hot element, butane torch, and lighter ignition with a propellant. The initial tests were conducted when fuel moisture levels were too high (>200%, expressed as a percentage of dry weight) to allow self-sustaining combustion. As the fuel dried over time, RSC was self sustaining for several minutes, and yielded CO and $PM_{2.5}$ EFs that were similar to those observed in the field. During these tests, grab samples during predominantly flaming and smoldering phases were collected in Summa (Cinnaminson, NJ, USA) stainless steel canisters for trace gas analysis using EPA Methods TO-15. Target compounds in the gas and PM phase include saturated (alkane) and unsaturated hydrocarbons, aldehydes, ketones, organic acids, and polycyclic aromatic hydrocarbons (PAHs). Levoglucosan in $PM_{2.5}$ is presented for the smoldering soil smoke since it is a commonly used marker for biomass combustion. Continuous total hydrocarbon measurements were also made using EPA Method 25A and a TECO Model 51 THC Analyzer which employs flame ionization detection with no chromatographic separation. Dinitrophenylhydrazine (DNPH) cartridges (LabCorp, Cary, NC, USA) were used to quantify carbonyl emissions (EPA method TO-11A). PM_{10} and $PM_{2.5}$ measurements were made using inertial impaction gravimetric filter techniques (EPA Method IP-10A) upstream from the canisters. These samples were then subjected to particle and total gaseous carbon analysis using thermogravimetric analysis and gas chromatography (GC) using an Agilent (Santa Clara, CA, USA) Model 6890 GC and Model 5975 mass spectrometer (methods are described in detail in Geron and Arnts, 2010). The impactors were backed by polyurethane foam traps for collection of gas phase semivolatile organic compounds that pass through or are volatilized from the filters (EPA Method TO-10A, see complete sampling and analytical protocol in Hays et al., 2002). Total emissions are then determined by multiplying the EF by the dry weight of total fuel consumed as discussed below.

CO_2 was analyzed via infrared gas techniques (California Analytical Model ZRH CO_2 Analyzer, Orange, CA, USA, and LICOR Model 840, LICOR, Inc. Lincoln, NE, USA) to account for CO_2 carbon in the mass balance flux techniques and to characterize the nature of plume dispersion and proximity to the combustion zone. We used the $CO/CO_2/VOC$ measurements to help us in chemically identifying the flaming and smoldering stages of the fires in addition to visual assessment.

CO was monitored continuously with EPA Method 10 using gas filter correlation and infrared detection (Thermo Electron Model 48C Thermo Environmental Instruments, Inc., Franklin, MA). The portable continuous CO (H_2SO_4 reaction) detector was also employed as discussed above. Standard gases (100 and 600 ppm,

Scott Specialty Gases, Plumsteadsville, PA) were used to calibrate the CO analyzers.

SO_2 measurements were performed with EPA Method 6C (pulsed chemical fluorescence) using a Thermo Environmental Instruments, Inc, Model 43S (Franklin, MA, USA). NO_x measurements were performed with EPA Method 7E (chemiluminescence) using a Thermo Environmental Instruments, Inc, Model 42S (Franklin, MA, USA).

Background measurements of PM and trace gases were performed by air monitoring prior to fire ignition and again after burning ceased or the inlets were moved away from the burning area. CO_2 and CO background concentration values were subtracted from values observed in smoke samples prior to EF calculations. NIST-traceable certified span gases (Scott Specialty Gases, Inc., Plumsteadsville, PA, USA) were used to perform five-point calibrations of the continuous emission monitors on site prior to monitoring. Serial dilution was performed using a dynamic dilution system (Model 146, Thermo Environmental Instruments, Inc., Franklin, MA, USA).

4. Emissions factor calculations

Landscape scale emissions of trace gases and PM are typically determined using the approach of Taylor and Zimmerman (1991) and Hao and Liu (1994):

$$M = A \cdot B \cdot \alpha \cdot \beta \tag{1}$$

where M is the amount of biomass consumed annually (dry tonnes yr^{-1}), A is the total land area burned annually (ha yr^{-1}), B is the average organic matter (fuel load) per unit area in individual biomes (metric tonnes or MT ha^{-1}), α is the fraction of above ground biomass relative to total (adjusted here to include organic soils), and β is the burning efficiency (fraction consumed) of the above-ground biomass. Total emission of a given compound is calculated by multiplying M by an emission factor, which is typically expressed in units of g kg^{-1} dry fuel consumed.

These emission factors and total emissions of trace gases and PM from individual fires are typically determined using a carbon mass balance approach as described in Ward et al. (1988). Using the same basic approach, we estimate emission factors as:

$$EF_t = \frac{C_t}{(C_{CO_2} + C_{CO} + C_{CH_4} + C_{TPM} + C_{VOC})F_C} \tag{2}$$

where EF_t is the emission factor (g kg^{-1} fuel dry weight) of the target compound(s), C_t is the concentration (g m^{-3}) of the target compound(s), $C_{CO_2,CO,CH_4,TPM,VOC}$ are sample C concentrations of CO_2, CO, CH_4, total PM, and total VOC, respectively, and F_C is fraction of carbon in the dry fuel mass. This is approximated here as 0.54 and is discussed below. The C concentrations used here are corrected by subtracting concentrations of the carbon constituents in the background as determined by measurements in ambient air before and after sampling of smoke plumes at the respective sites.

Emission fluxes are then determined from

$$F_t = EF_t \times CS_F \tag{3}$$

where F_t is the flux of the target compound(s) in kg ha^{-1} and CS_F is the fuel consumption (dry tonnes ha^{-1}). Nitrogenous emissions are calculated similarly. We used this approach to calculate EFs and total emission fluxes from the coastal plain smoldering soil and above ground prescribed fires. We include CH_4 in the total VOC estimates, since CH_4 was not measured independently in all samples. In this paper, the primary focus is on the NAAQS criteria pollutants CO and $PM_{2.5}$.

NO_x and SO_2 emissions were estimated for the laboratory tests and above ground fuels and are discussed in a separate study. EFs for those compounds were found to fall within ranges summarized by Andreae and Merlet (2001).

5. Results and discussion

Above-ground fuel consumption from the prescribed burns described here generally falls within the range of values (3–20 tons acre^{-1} or 6.8–45 tonnes ha^{-1}) assumed in current models and smoke management guidelines (North Carolina Smoke Management Program, 2008). In cases where fine fuels (such as marsh grass or pine litter) accumulate, fuel loads range from 3 to 6 tons acre^{-1} (6.8 to 13.5 tonnes ha^{-1}). Where flammable shrubs (such as gallberry and fetterbush) colonize sites, fuel loads and consumption may be considerably higher. These circumstances occur when fire frequency is reduced and fuels are allowed to accumulate, creating wildfire risks. Wildfire fuel consumption was difficult to estimate, but was much higher, since larger fuels were typically consumed. Where soil fires occur, they can burn to a depth of a meter or more, resulting in ~2000 (1000–6000) tonnes ha^{-1} of organic soil consumption. This value is at least an order of magnitude greater than above ground fuel consumption. Organic soil bulk density ranges from 0.1 to 1.1 g cm^{-3} (Lindsay, 2010; Hashim and Islam, 2008), and a range of 0.1–0.6 g cm^{-3} was used to derive the fuel consumption range above. Values for the U.S. coastal plain are usually less than 0.5 g cm^{-3}, although the woody peat composing the organic soils at PLNWR was determined to be of slightly higher bulk density. Therefore, soil fuel consumption on this site may fall at the upper end of this range.

In general, our PM$_{2.5}$ EFs from above ground fuels from prescribed burning are in agreement with those published in AP-42 (U.S. EPA, http://www.epa.gov/ttnchie1/ap42/) and those summarized by Vose et al. (1997). The PM$_{2.5}$ EFs for coniferous and mixed fine fuels span the range of those used in the emission model (6–12 g kg^{-1}) described by Wiedinmyer et al. (2006) for flaming combustion in coniferous, mixed, and shrubland fuel classes. The PM$_{2.5}$ EF of 8.0 g kg^{-1} used by Dennis et al. (2002) for litter and fine woody fuels is similar to the values observed from our forest prescribed burns dominated by litter and duff consumption.

On the other hand, we found the CO and PM$_{2.5}$ EFs from smoldering soil combustion at PLNWR, Green Swamp, and ARNWR to be considerably higher (and CO$_2$ EFs lower) than previously reported

from above-ground biomass burning fuels. The combination of higher PM$_{2.5}$ and CO EFs and lower CO$_2$ EFs indicates that the organic soil fires exhibited more inefficient combustion than above-ground burns. The difference is reflected in Figs. 1 and 2, where the modified combustion efficiency (MCE) is plotted for prescribed (Green Swamp) and smoldering soil (Green Swamp and ARNWR) fires. MCE is calculated as

$$MCE = \Delta CO_2/(\Delta CO_2 + \Delta CO) \qquad (4)$$

where ΔCO_2 and ΔCO are the concentrations of CO$_2$ and CO in smoke minus their respective ambient background concentrations. MCE is much lower for the smoldering soil fires compared to the prescribed burn at Green Swamp (Figs. 1 and 2). MCE values for the soil fires are at the low end of the range reported for smoldering fires by Akagi et al. (2010), probably due to low aeration and oxygen availability during consumption of soil organic layers below the duff layer. Combustion of the soil and large diameter fuel components often results in higher emission of products of incomplete combustion (McMahon et al., 1980; Bertschi et al., 2003). Our OC/TC values also suggest very inefficient combustion in the field at PLNWR and ARNWR, with values exceeding 0.98. This is consistent with high OC contents of PM$_{2.5}$ previously reported in plumes from these fires (Geron, 2009, and observations made later at that site). Combustion of above ground fuels typically yield OC/TC values ranging from 0.8 to 0.9 (Andreae and Merlet, 2001).

CO$_2$ and CO concentrations and MCE (Fig. 1) from the ARNWR suggest a lower CO EF and somewhat higher CO$_2$ EF derived from plumes further away from biomass combustion sources. The CO/CO$_2$ values are higher closer to the ground fire sources, suggesting CO EFs calculated from concentrations in the aged and diluted smoke are somewhat lower than those calculated from the concentrations near the source. However, this difference may be due to more efficient combustion from the sources of the plumes sampled earlier in the day, although no flaming combustion was observed. PM$_{2.5}$ EFs do not seem to show this effect over this range in distances (Table 1), possibly due to condensation onto existing particles and particle growth in the more aged smoke.

Previous studies of organic soil burning PM EFs are few and mostly limited to laboratory investigations. Benner (1977) reported total suspended particulate (TSP) values of 44 ± 9 g kg^{-1}. These measurements were determined from 1 g peat samples where combustion was sustained with a hot wire heat source.
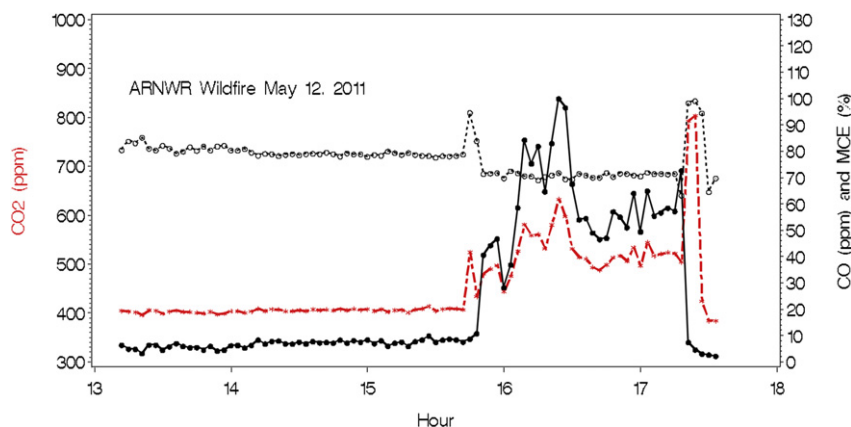

Fig. 1. CO (black dots, solid line), CO$_2$ (red stars, dashed line) concentrations and MCE (black circles, dotted line) from smoke (plus background) approximately 100 m from the source (before 15:45) and within 5–10 m of the source (after 15:45) at ARNWR. CO and CO$_2$ measurements were logged at two second intervals, and are shown here as three minute averages. Smoke was predominately from smoldering ground fire. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
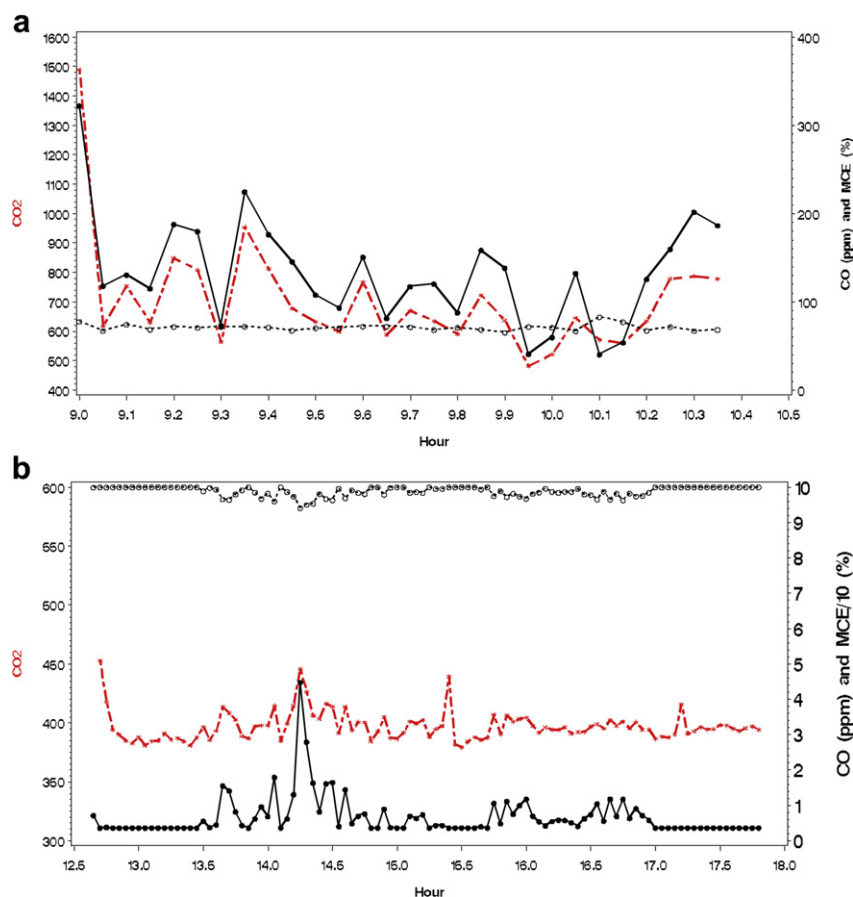
Fig. 2. a) CO (black dots, solid line), CO_2 (red stars, dashed line) concentrations and MCE (black circles, dotted line) from smoke (plus background) approximately 1 m from the source at Green Swamp. Smoke was from smoldering ground fire. MCE ranged from 65 to 80%. b) CO (black dots, solid line), CO_2 (red stars, dashed line) concentrations and MCE (black circles, dotted line) from smoke (plus background) approximately 50–100 m from the source at Green Swamp. Smoke was from predominantly flaming prescribed fire. MCE ranged from 94 to 99%. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

McMahon et al. (1980) also found that PM EFs can exceed 40 g kg^{-1} (30 ± 20 g kg^{-1}) from Florida sawgrass peat. Ward (1990), and Ward et al. (1988) reported a TSP EF of 35 g kg^{-1} from 30% organic soil mixed with bulldozer piled logging slash.

Table 1
Wildfire emission factors (g kg^{-1} fuel dry weight consumed) from field samples collected during this study compared with laboratory values previously published. N is the number of filter samples taken for PM$_{2.5}$ mass determination.

	PM$_{2.5}$	N	CO$_2$	CO	VOC
ALNWR 2011					
Ground fire	48–66	4	1092–1390	220–290	21–49
Mixed/Aged	55–79	4	1120–1440	125–161	17–29
Green Swamp 2009					
Ground fire	44–53	4	1100–1140	260–280	30–40
Prescribed fire	9–16	4	1450–1640	10–80	10–33
PLNWR 2008					
Ground fire	34–55	3	1010–1140	230–300	18–38
Previous studies[a]					
Benner (1977)	44 ± 9[b,c]			251 ± 110[b]	52 ± 31[b]
Chen et al. (2007)	41.7		2784[d]	271	125
Christian et al. (2003)			1703	210	76[e]
McMahon et al. (1980)	30 ± 20[b,c]			269 ± 130[b,f]	23 ± 18[b,f]

[a] Laboratory derived emission factors from peat/organic soil combustion.
[b] Standard deviation.
[c] Total suspended particulate (TSP).
[d] Expressed in g kgC^{-1}.
[e] From Table 6, includes methane.
[f] Mean of wet and dry organic soils.

Chen et al. (2007) reported an EF of 41.7 g kg^{-1} from Alaskan tundra organic soil cores.

Although PM sizes were not measured in the laboratory studies cited above, pyrolysis studies of peat have previously shown aerodynamic diameters to be less than 1 μm (Skiotis et al., 1973). Similarly, the PM$_{10}$ results were not significantly different from the PM$_{2.5}$ data presented here, so we did not include PM$_{10}$ estimates separately. Two matching pairs of PM$_{10}$ and PM$_{2.5}$ from the Green Swamp and ARNWR field soil fires were within 10%. The laboratory results from the peat cores were within 3% on average and one PM$_{10}$ mass was actually slightly less than PM$_{2.5}$, so for that pair the measurement error was likely greater than the true difference between PM$_{10}$ and PM$_{2.5}$ emissions. The soil burning PM$_{2.5}$ EFs at PLNWR, Green Swamp, and ARNWR exceeded 40 g kg^{-1}, and the laboratory studies of the peat fuels from PLNWR also exceeded this value (Table 1).

The laboratory studies of organic soil combustion performed here were conducted as a means to isolate the peat fuel soil source and better understand conditions controlling ignition and PM$_{2.5}$ emission. Soil organic matter harvested from the PLNWR pine plantation where ground fire occurred in 2009 was used for these tests. Proximate/ultimate analysis of the peat cores indicated that the dry composition was 54–60% carbon; 20–21% oxygen; 13–17% ash; 4.9–5.3% hydrogen; 0.3–1.6% sulfur; and 0.9–1.0% nitrogen. Total fuel carbon by mass typically ranges from 45 to 54% of the fuel dry weight for most biomass fuels. The carbon content of the woody peat from PLNWR was found to be somewhat higher (54–60%) here,

but this range is consistent with peat values from Christian et al. (2003). Woody fibers composed a substantial portion of the peat at PLNWR, while organic muck soils were more prevalent on the ARNWR. A series of ignitions was attempted as the fuel dried over a period of several days. The organic soil samples would not sustain combustion until moisture declined from over 200% to less than 30%. $PM_{2.5}$ emissions ranged from 50 to 150 g kg^{-1}, while CO EFs ranged from 220 to 340 g kg^{-1}. Formaldehyde, methane, and total organic compound EFs also exceeded values published for above ground fuels but were highly variable.

Carbon monoxide EFs from the *in situ* and laboratory soil burns were higher than those reported from above ground fuels by approximately a factor of three, and are consistent with a few previous laboratory reports (Table 1). Benner (1977) reported CO EF values of 251 ± 110 g kg^{-1}. McMahon et al. (1980) reported a soil burning CO EF of 269 g kg^{-1}, while Chen et al. (2007) also report a tundra soil core CO EF of 271 g kg^{-1}, all of which fall within our range of 220–340 g kg^{-1}. Christian et al. (2003) report a CO EF of 210 g kg^{-1} from Indonesian peat fuels. Typical CO EF values from other (above ground) biomass burning fuels are less than 100 g kg^{-1}, with a mean of approximately 80 g kg^{-1} (Andreae and Merlet, 2001). This is consistent with the values we observe during the Green Swamp prescribed fire.

Levoglucosan is a tarry sugar anhydride derived from cellulose combustion and is commonly used as a tracer of biomass burning particulate (Hays et al., 2002). Sabiham (2010) found that peats contained less than 10% cellulose, while 20–50% of leaf litter, wood, and other living plant biomass (dry weight) is composed of cellulose, primarily in cell walls (Salisbury and Ross, 1978). We suspected that lower cellulose fractions in the organic soils would lead to lower values of levoglucosan in $PM_{2.5}$ from organic soil fires compared to burning of fine above-ground fuels. However, levoglucosan analysis using the methods of Hays et al. (2002) indicated that similar fractions of levoglucosan (2–3%) were found in $PM_{2.5}$ from smoldering organic soil fires at PLNWR where samples were collected in August 2008 during the Evans Road Wildfire from a deep smoldering peat seam. Fabbri et al. (2009) found similar levoglucosan levels in $PM_{2.5}$ from lignite or "brown" coals, where pyrolysis yielded $PM_{2.5}$ composed of 1.6–3.5% levoglucosan. They also reported that galactosan was absent from lignite $PM_{2.5}$, and mannosan was found in much lower concentrations than reported in biomass smoke. These findings suggest that levoglucosan may not be used to distinguish above ground biomass burning $PM_{2.5}$ from that of organic soil or brown coal, but other sugar derivatives may be used for this distinction instead.

Little data on emission fluxes from southeastern U.S. organic soil fires is currently available, and this fire type in particular is projected to increase in the future with changing climate and land management practices such as drainage for agriculture (Page et al., 2002). Our data, when examined in the context of previous studies, suggest that emissions of $PM_{2.5}$, CO, and likely other gas phase reduced compounds (many of which are air toxics) will be lower during prescribed fires compared to wildfires covering the same area. This is largely because wildfires typically occur during excessively dry periods, when organic forest soils and heavy debris are dry and susceptible to smoldering/incomplete combustion, the source of many toxic compounds. These wildfires may increase fuel consumption by an order of magnitude or more since large quantities (100s–1000s tonnes ha^{-1}) of soil and heavy fuels can be consumed under these dry conditions. For instance, Page et al. (2002) found that over 500 tonnes ha^{-1} of organic soil could be consumed in tropical peatland fires. In addition, our data and those from Benner (1977), McMahon et al. (1980), Bertschi et al. (2003), and Chen et al. (2007) indicate that EFs for PM and reduced trace gases may be 2 to over 10 times higher from these soils and heavy

fuels compared to the lighter fuels typically consumed during prescribed burns.

The Evans Road wildfire (on the PLNWR) during spring and summer of 2008 released an estimated 9.4 Tg or million metric tons of carbon (Mickler et al., submitted for publication). Of this carbon release, 98% was estimated to evolve from below-ground fuels. If we apply a $PM_{2.5}$ EF of 40 g kg^{-1} and 50% C fraction to the organic soils, a total $PM_{2.5}$ emission of 730,000 metric tons (0.73 Tg) is derived, which is over 17% of $PM_{2.5}$ emissions from all U.S. sources (EPA TRENDS, 2012, where fire emissions are calculated primarily from consumption of above-ground fuels) and over twice the 2008 estimated total $PM_{2.5}$ emissions for North Carolina. This peat wildfire episode was also found to be associated with both respiratory and cardiac effects within the exposed human population in eastern North Carolina (Rappold et al., 2011). Similar calculations for the 2011 Pains Bay Fire (on the ARNWR) have a similar air quality impact. It is estimated that soil was consumed on roughly half of the burned area to a depth ranging from 15 to 30 cm, with some deeper pockets near the base of trees. This was observed on four transects through the ARNWR, and was supported by the observations of Tom Crews (Fire Management Officer, U.S. Fish and Wildlife Service, Alligator River NWR) and Ed Christopher (Fire Management Officer, Pocosin Lakes NWR). A mean soil burn depth of 20 cm yields a total $PM_{2.5}$ emission of 440,000 metric tons (0.44 Tg), which is over 10% of $PM_{2.5}$ emissions from all U.S. sources (EPA TRENDS, 2012). Applying a CO EF of 250 g kg^{-1} yields total CO fluxes of 4.7 and 2.75 Tg from the PLNWR and ARNWR, respectively. This represents 4–7% of the national total CO emission inventory, and 25–42% of the estimated above-ground national wildfire CO emissions.

The uncertainty in emission factors and fuel consumed suggest an overall uncertainty of at least 50% in the total $PM_{2.5}$ and CO emission estimates from these fires. The EFs used in these estimates, while consistent with laboratory measurements, may be at the low end of actual field rates given the range we observed in the field. On global scales, large fires in tropical (Page et al., 2002) and boreal (Shvidenko et al., 2011) peat systems can have a significant impact on global carbon budgets as well.

Amounts of smoke from peat fires may therefore be 100–500 times higher per unit burned area than emissions from prescribed burns. Peat fires occur frequently on the southeastern U.S. coastal plain, but their seasonal and annual extent varies widely. Reardon et al. (2009, and references therein) documents fire return intervals of 13–50 years for low pocosins with soils high in organic content. However, consumption of the soil itself appears to be driven by hydroperiod, with dryer soil profiles leading to greater organic soil consumption. These authors infer annual extent of soil fires to be on the order of a few thousand hectares on the coastal plain. On the other hand, six large wildfires on the southeastern coastal plain have occurred since 2007 that have consumed nearly 300,000 ha. This area was dominated by fires in North Carolina and the 2007 fire in the Okefenokee refuge on the Georgia-Florida border.

Plume rise due to lower heat release rate from smoldering soil combustion itself is no doubt slower. However, when these plumes are entrained into daytime turbulent convectively mixed boundary layers, they are often transported hundreds of kilometers. In fact, smoke from the Okefenokee, PLNWR and ARNWR ground fires severely impacted air quality ($PM_{2.5}$, CO) in the Raleigh/Durham area hundreds of kilometers away in 2007 and 2008 (Geron, 2009) and again in 2011. In addition, human exposure to fire and smoke is likely to be much greater during wildfires since ventilation, wind conditions, and other factors influencing smoke dispersion cannot be coordinated with wildfire events and especially long term smoldering soil fires, as they can with prescribed burning. Fires in

southeastern peat soils also may smolder beneath the surface and re-ignite without detection. These subsurface fires are dangerous to firefighters and others present on the landscape. In addition, long-term ecosystem damage may result from these wildfires. In the Southeastern U.S. wildfires can cause damage to red cockaded woodpecker (RCW) nest trees, habitat which can take many decades to restore. This habitat has been reduced by over 97% since the 1700s (USDA, 1988) due to land management practices, placing the RCW and other species such as the gopher tortoise in endangered status. Prescribed fire use and other strategies are needed to reduce 1) human exposure to and emissions of hazardous air pollutants, 2) net risk to property and human welfare, and 3) damage to critical (RT&E) wildlife habitat by reducing wildfire hazards. Additional field studies to collect additional (*in situ*) ground fire emission data from ecosystems susceptible to fire are needed. Currently available EFs for soil/peat fuels are largely limited to a few laboratory observations. Fuel consumption estimates and knowledge of smoke transport characteristics associated with organic soil fires is also limited or non-existent and is needed for air quality modeling exercises to support exposure and policy analysis.

Acknowledgments

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