Particulate emissions from different types of biomass burning

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HIGHLIGHTS

► Chemical speciation of $PM_{2.5}$ emissions shows differences among different types of biomass burning.
► Emissions of polar organic compounds depend on fuel types during biomass burning.
► Combination of chemical tracers allows to characterize prescribed burning emissions.

ARTICLE INFO

ABSTRACT

Biomass burning is a significant emission source of $PM_{2.5}$ (i.e., particulate matter with an aerodynamic diameter less than 2.5 μm), but few studies addressed the chemical composition of $PM_{2.5}$ emissions from various types of fires. Here, we present results from a sampling campaign to quantify $PM_{2.5}$ emissions from various types of prescribed burning activities using analysis of carbon (elemental carbon: EC; organic carbon: OC; and total carbon: TC); polar organic compounds (12 different compounds and four functional classes); water-soluble potassium (K^+); and particle-bound mercury (PHg). Emissions were characterized for a series of prescribed burns in the Lake Tahoe basin in the western United States, along with controlled biomass combustion in a wood stove. In the field, emissions were collected from: (i) *landscape underburns*, consisting of wooden tissues, foliage, branches, and surface duff; (ii) *pile burns*, consisting mainly of wooden tissues stacked up to piles; (iii) *mixed underburn/pile* burns which consisted of a mix of the above; in a wood stove, burns included different fuel types collected from the Lake Tahoe basin, specifically (iv) wooden logs mainly of pine; (v) green foliage and branches from two dominant shrubs (manzanita and bitterbrush); and (vi) surface duff, mostly consisting of pine needle litter.

Our data showed higher ratios of organic to elemental carbon in green fuels (19.2 ± 4.2) compared to dry, wooden logs (7.3 ± 1.9) both in prescribed burns in the field and in controlled stove combustion, indicating that more moisture in green biomass resulted in more smoldering-phase combustion. Further, OC/EC ratios were lower in wood stove burns compared to prescribed burns in the field, which we attribute to higher combustion temperatures in wood stove burns. The suite of 12 select polar organic compounds showed that the most prevalent compounds emitted across all burns were levoglucosan, mannosan, and resin acids (dehydroabietic, pimaric, and abietic acids), while emissions of inositols and arabitols were only significant in combustion of leaves from a broadleaf shrub indicating their potential use as tracers for green foliage. Water-soluble K^+, a common tracer for biomass combustion, showed a clear difference between field understory burns (low K^+) and wooden pile burns (nearly 5 times higher), suggesting that K^+ can potentially be used for differentiating between different prescribed burning types. Finally, PHg emissions were lowest in green vegetation and underburns emissions, which was unexpected due to inherently higher Hg levels in green foliage and surface duff. Using multiple tracers (i.e., soluble potassium, carbon and mercury) allowed to separate emissions between prescribed burning, controlled stove burning, and ambient air which includes residential wood combustion.

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1. Introduction

Biomass burning (including forest fires, agricultural waste burning and residential wood combustion) contributes more than one third of primary $PM_{2.5}$ (i.e., particulate matter [PM] with an aerodynamic diameter less than 2.5 μm) loads in the U.S. (Chen

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et al., 2007). PM$_{2.5}$ pollution leads to reductions in atmospheric visibility (Green et al., 2012) and can cause acute health effects (Huttunen et al., 2012). The relative importance of PM emissions from various biomass combustion types, such as from wildfires, prescribed fires, or residential wood combustion, is poorly quantified (Kuhns et al., 2004; Urbanski et al., 2011). In the U.S., research has focused on characterizing smoke emissions from wildfires (Strand et al., 2011; Sullivan et al., 2008; Ward et al., 2006); fewer studies have addressed emissions from prescribed burning (Hardy et al., 1996; Lee et al., 2005; Mazzoleni et al., 2007; Sullivan et al., 2008; Chen et al., 2010; Shackelford, 2010).

One area with a significant amount of biomass burning from both prescribed burning and fireplace/woodstove activities is the Lake Tahoe basin. Lake Tahoe is the second deepest lake in U.S. and with a depth of over 500 m, this pristine, alpine lake is well known for its excellent water clarity. Various environmental concerns in the basin are related to water quality and lake clarity. For example, Secchi depth, a measure of water clarity, has dropped from 31 m in 1968 to less than 20 m in 2010, believed to be mainly caused by inputs of nutrients (nitrogen and phosphorus) and fine particulates to the lake (UCDAVIS, 2011). To reduce PM pollution in the Lake Tahoe basin, it is necessary to know the contributions and chemical composition of various emission sources, including different types of biomass burning activities that are present in the basin. Studies indicate that residential wood burning, along with impacts from local and regional wildfires, is a potentially significant source for PM$_{2.5}$ during the fall and winter (Engelbrecht et al., 2009). Chemical mass balance results, based on two south-shore sites in the basin, further indicate that between 51 and 67% of PM$_{2.5}$ may originate from wood burning (Engelbrecht et al., 2009).

Prescribed burning has become a common strategy to reduce the risk of wildfires as controlled combustion of fuels has shown to reduce the risk for wildfires. For example, prescribed fires reduced fuel mass by 67%–88% in a Sierra Nevada mixed conifer forest (Knapp et al., 2005). A recent study showed that reduction in fuel loads by prescribed fires led to lower CO$_2$ emissions during the occurrence of subsequent wildfires by 18–25% in the western U.S., and up to 60% in other forest systems (Wiedinmyer and Hurteau, 2010). In the Lake Tahoe basin, over 3000 acres of pile burning and 1000 acres of landscape underburns have been conducted since 1997 (USDA, 2009). Prescribed burning potentially contributes to declining air and water quality by releasing nutrients and particulates (Shackelford, 2010).

In order to develop potential tracers for the areas affected by various types of biomass combustion, we characterized PM$_{2.5}$ emission composition of various biomass combustion types, including field and laboratory combustion. In the field, we sampled emissions from: (i) *landscape underburns*, consisting of wooden tissues, foliage, branches, and surface duff; (ii) *pile burns*, consisting mainly of wooden tissues stacked up to piles of 0.2–0.3 m diameter; (iii) *mixed underburn/pile* burns which consisted of a mix of the above. In a wood stove at our institute, we conducted controlled combustion of biomass collected from the Lake Tahoe basin, including (iv) wooden logs (mainly pine); (v) green foliage and branches from two dominant shrubs (manzanita and bitterbrush); and (vi) surface duff, mostly consisting of pine needle litter. Finally, we also conducted ambient-air sampling in residential neighborhoods during seasons when strong emissions from domestic wood combustion was expected (i.e., winter months). To characterize chemical composition of PM$_{2.5}$ emissions, we measured the following chemical constituents: organic carbon (OC); elemental carbon (EC); water-soluble potassium (K$^+$), a common tracer for biomass burning emissions; a suite of 12 different polar organic compounds considered characteristic of biomass burning (Burshtein et al., 2011; Mazzoleni et al., 2007; Medeiros and

Simoneit, 2008; Simoneit and Elias, 2000; Simoneit et al., 1993); and particulate-bound mercury, a compound that may be used to differentiate various fuel sources because of inherently different Hg levels of different tissues (Obrist et al., 2009, 2011).

2. Material and methods

2.1. Sampling sites

Vegetation in the basin is characterized as mixed conifer forests, dominated by Jeffrey pine (*Pinus jeffreyi* Grev. & Balf. [Pinaceae]), Sierra lodgepole pine (Pinus contorta var. murrayana [Grev. & Balf.] Engelm.), white fir (*Abies concolor* [Gord.& Glend.] Lindl. ex Hildebr. [Pinaceae]), and California red fir (Abies magnifica A. Murr.). The basin also contains large areas of shrub understory, including greenleaf manzanita (*Arctostaphylos patula* Greene [Ericaceae]) and desert bitterbrush (*Purshia tridentata*[Pursh] DC [Rosaceae]).

Samples of smoke emissions from prescribed burning activities were collected in spring, winter, and fall in 2010 and 2011 from several areas in the Lake Tahoe basin. Field sites included: (i) Sugar Pine State Park on the west side of Lake Tahoe; (ii) Pioneer Trail located in South Lake Tahoe; (iii) Tunnel Creek on the north–east side of the lake; and (iv) Rubicon Bay on the southwest side of Lake Tahoe. To conduct controlled combustion experiments in a wood stove at the Desert Research Institute in Reno, NV, USA, we purchased two types of dried wooden logs from a local grocery store (i.e.Pine/cedar/Fir logs and Jeffery Pine logs), collected green foliage and branches in the basin composed of approximately 95% manzanita and 5% Bitterbrush, and sampled surface litter. Finally, ambient air sampling was conducted in two residential areas in the basin; in Tahoe City from December 30, 2010 to January 1, 2011 and from January 4 to January 5, 2011; in Incline Village from December 6 to 8 and December 21 to 23, 2011.

2.2. Sampling procedure

For sampling, medium volume samplers were placed as close to the fires as possible; sampling distances to the fires ranged from 20 m to 200 m from the fire source. Samplers consisted of an inlet cyclone (Model TE-1008-9-2.5, Tisch Environmental, Ohio, USA), a vacuum pump (Model 1023, Gast Manufacturing Inc., Michigan, USA), a mass flow control valve, and a mass flow meter (Model 4140, TSI, Shoreview,MN, USA). Two identical samplers were run in parallel to collect two filter samples and to allow for enough sample material for analyses of all chemical compounds. 100-mm diameter quartz fiber filters (Pall Life Sciences, Ann Arbor, MI, USA) were used for sample collection. In general, filters were sampled for about 30 min to 2 h during prescribed burning, while filters were sampled for 12-h periods during ambient air sampling (separated into night and daytime). All filters were heat-treated prior to filter sampling at 900 °C for at least 4 h for cleaning. Flow rates were set at 113 L min^{-1} during sampling.

2.3. Analytical procedures

Each sampled filter was divided for analysis: half a filter was used for mercury analysis, and each one quarter of a filter was used for carbon and K$^+$ analysis. Half of the second filter was used for analysis of polar organic compounds and the last half was kept for a backup for repeated analysis. OC and EC were determined by the Interagency Monitoring of Protected Visual Environments (IMPROVE)_A thermal/optical reflectance (TOR) protocol using a DRI model 2001 carbon analyzer (Atmoslytic, Inc., Calabasas, CA, USA)(Chow et al., 1993). OC was separated into OC1 to OC4 plus pyrolyzed OC, and EC equaled fractions EC1 to EC3 minus pyrolyzed

OC(Chow et al., 2001). The detection limits for OC, EC, and TC are 0.39 µgC cm^{-2}, 0.01 µgC cm^{-2}, and 0.42 µgC cm^{-2}, respectively, and relative standard deviations of replicate analyses were 2–4%. Sample filter loadings were at least 45, 298 and 52 times higher than these detection limits for OC, EC and TC respectively.

A quarter filter was extracted in tubes of 15 ml of distilled deionized water. The extraction tubes were then sonicated for one hour, shaken for one hour, and refrigerated until analysis. Water-soluble potassium was analyzed by Atomic Absorption Spectroscopy (Grover et al., 2006) using a Varian SpectraAA 880 (Varian Australia Pty Ltd, Mulgrave, Victoria, Australia). The detection limit for soluble K$^+$ was 0.010 µg ml^{-1} and relative standard deviations of replicate analyses were 2–5%.The quartz filter blank and field blank for soluble potassium averaged 0.035 ± 0.010 and 0.050 ± 0.011 µg ml^{-1}. Soluble K$^+$ of filter blanks and field blanks were below 2% of loadings on sample filters.

Polar organic compounds were extracted by accelerated solvent extraction (Dionex 300 Accelerated Solvent Extrator) with 80% methanol and 20% water at 80 °C and 1500 psi. After extracting them twice with the solvent mix (total volume of solvent was ~100 ml), extracts were concentrated by rotary evaporation to a final volume of 40 ml. A 500 µl sample was taken and dried to eliminate water and methanol that would later interact with derivatizing reagents. 50 µl of acetonitrile was added, followed by 50 µl of pyridine and 150 µl of BSTFA with 1% TMCS [N,O-bis-(trimethylsilyl) trifluoroacetamide with 1% trimethylchlorosilane (Thermo Scientific, Waltham, MA, USA)]. These derivatizing reagents convert polar organic compounds into their trimethylsilyl derivatives for analysis of organic acids, anhydrosugars (levoglucosan and mannosan), and sugar alcohols (arabitols, inositol, etc.). For the derivatization reaction, vials (Waters Corporation, Milford, MA, USA) containing samples were placed onto a thermal plate heated to 65 °C for 2 h. Calibration solutions were made fresh in methanol and derivatized just prior to the analysis of each sample set. Samples were analyzed by gas chromatography interfaced with mass spectrometry (GC/MS) within 18 h to avoid degradation. The samples were analyzed by an electron impact ionization GC/MS technique using a Varian 4000 gas chromatograph mass spectrometer (Varian, Inc., Walnut Creek, CA, USA).

Total mercury was measured after acid digestion using a Tekran Model 2600 water analyzer according to method 1631 revision E. Briefly, half of a quartz filter was digested by 10% (v/v) bromine monochloride (BrCl) solution for 12 h under laboratory conditions. Prior to analysis, pre-purified hydroxylamine hydrochloride was used to neutralize excess BrCl. For analysis, samples were mixed with stannous chloride (SnCl$_2$) in a phase separator, whereby oxidized Hg was reduced to volatile elemental Hg which was loaded onto two sequential gold traps by means of Argon carrier gas. After thermal desorption, elemental Hg was detected by atomic fluorescence spectrometry. The Tekran Model 2600 was calibrated using NIST SRM-3133 Hg standards (0, 0.5, 1.0, 5.0, 10.0, 25.0, and 50.0 ng l^{-1}Hg). The detection limits (DL) was estimated at about 3 ng l^{-1}. Ongoing Precision Recovery (OPR; 5 ng l^{-1}) was periodically conducted to check the reliability of the system, and averaged 103.2% ± 6.8%. All samples were analyzed in duplicate.

2.4. Statistical analyses

Statistical tests were performed using the statistics analysis software STATA (Version 9.1, Stata Corporation, College Station, Texas, USA). Analysis of variance (ANOVA) and Bonferroni post-hoc comparison tests were run to analyze for statistical effects of variables: burning environments (ambient air vs. field vs. controlled stove) and fuel types (wooden logs vs. green foliage vs. litter/duff). Dependent variables tested included OC/EC ratios, percentage

contributions of the four groups of polar organic compounds, water-soluble potassium TC^{-1} ratios, and PHg TC^{-1} ratios.

3. Results and discussion

3.1. OC/EC ratios

OC/EC ratios are shown in Table 1 and Fig. 1A. In the field, OC/EC ratios were highest in underburns, and were lower in pile burns and mixed pile/underburns which were dominated by wooden logs. Emissions from controlled biomass combustion in a wood stove similarly showed lower OC/EC ratios during burning of wooden logs compared to green leaves, branches, and surface duff. Highest OC/EC ratios of all samples were observed in ambient air both in 2010 and 2011. OC/EC ratios from field prescribed burning were generally in the range of other studies that reported prescribed burning emissions in pine forests (from Georgia: a mean of 15: Lee et al., 2005; a mean of 32: Yan et al., 2008), but were higher compared to those from wildfires (e.g., a mean of 5.1; Pio et al., 2008). OC/EC ratios are often attributed to different fire intensity: higher temperature of flaming phase burnings have been shown to produce more soot (EC), and hence lower OC/EC ratios, than smoldering-type combustion (Pósfai et al., 2003). Our study supports the notion that fire intensity plays a role in OC/EC emission patterns as evident by generally lower OC/EC ratios when burning wooden logs (both in the field and in the controlled stove combustion) compared to combustion of greener fuel types (e.g., field underburns, and combustion of green leaves and branches in a stove). Yan et al. (2008) attributed higher OC/EC ratios in their study to lower flaming combustion during prescribed burns than in wildfires. Higher OC/EC ratios in prescribed fires are likely owed to lower combustion intensities during this type of burning (Robinson et al., 2011) as they are conducted when fuels are relatively moist and during periods when ambient air humidity is relatively high.

OC/EC ratios in ambient air samples were much higher than those of any biomass burning emissions. Ambient air OC/EC ratios (a mean of 26.3 and 38.4 for winter of 2010 and 2011 respectively) were also much higher than ratios observed in 174 rural sites of the IMPROVE network in the U.S from 2000 to 2004 (5; Zeng and Wang, 2011), but were similar to fine particle emissions from fireplace and woodstove combustion observed in remote Portuguese forests (24.1; Alves et al., 2011). Fine et al. (2001, 2002, 2004) also showed high OC/EC ratios (26) from fireplace combustion of softwood and hardwood. It is possible that high OC/EC ratios in ambient air of this study are co-determined by other sources; a previous source attribution study indicated that OC and EC from wood burning accounts for 15–74% of PM$_{2.5}$ mass in the Lake Tahoe basin (Kuhns et al., 2004). To our knowledge, however, many man-made emission sources show OC/EC ratios that are not nearly as high as those we observed in ambient air. For example, mobile sources (traffic) showed OC/EC ratios in the range of 1–23 for light-duty gasoline vehicles (Fujita et al., 2007) gas cooking showed ratios of about 5 from (See and Balasubramanian, 2008). On the other hand, McDonald et al. (2003) reported much higher OC/EC ratios from emissions of charbroiling of hamburger, steak and chicken under char (45, 35 and 360 on average respectively). Further, absorption of gas-phase volatile organic species onto the quartz fiber filter (Fine et al., 2001) may have contributed to high OC/EC ratios in ambient air samples in our study.

3.2. 12 Select polar organic compounds typical for biomass combustion

Total concentrations of compounds on the filters were extremely variable between different samples due to different

Table 1
Concentrations of organic carbon (OC), elemental carbon (EC), water-soluble potassium, 12 polar organics and particle-bound Hg; mean ± standard error.

Component	Units	Ambient air		Field burns			Stove burns		References			
		2010	2011	Pile burns	Underburns	Mixed pile/underburns	Logs	Leaves/duff	Ambient air	Prescribed burning	Fireplace/woodstove	Cooking
Organic carbon	mg C m⁻³	0.02 ± 0.00	0.02 ± 0.00	0.51 ± 0.14	4.78 ± 0.86	0.87 ± 0.30	17.73 ± 10.87	37.95 ± 5.08				
Elemental carbon	mg C m⁻³	<0.001	<0.001	0.04 ± 0.01	0.28 ± 0.09	0.07 ± 0.00	4.07 ± 2.72	4.57 ± 1.72				
OC/EC	mg C mg C⁻¹	26.2 ± 10.4	38.4 ± 12.0	13.0 ± 0.6	26.2 ± 7.1	12.2 ± 4.0	2.8 ± 1.3	12.1 ± 3.3	24.0[o]	15.4[b], 25.8[c], 88[d], 28.8[e], 20.7[f]	26.1[g], 42.2[h], 7.5[i], 3.1[j], 7.3[k], 119.3[l]	23.9[n]
K⁺ TC	%	0.8 ± 0.1	0.9 ± 0.2	1.2 ± 0.1	0.3 ± 0.1	1.2 ± 0.2	0.9 ± 0.1	1.6 ± 0.3		1.0[b],0.4[c], 0.5[d]	0.9[g], 1.1[i], 0.3[j], 0.7[k], 0.2[l]	3.10[n]
PHg TC⁻¹	ng g⁻¹	932 ± 75	1059 ± 229	156 ± 65	37 ± 13	189 ± 12	120 ± 21	7 ± 1				
Anhydrosugars TC	%	4.59 ± 0.47	0.69 ± 0.27	12.00 ± 2.03	8.93 ± 2.21	10.12 ± 1.76	3.62 ± 1.17	2.77 ± 0.31				
Levoglucosan TC	%	4.34 ± 0.43	0.67 ± 0.26	7.79 ± 2.57	8.05 ± 2.16	9.68 ± 1.57	2.51 ± 0.96	2.03 ± 0.34		8.9[b], 7.4[c], 6.3[e], 7.8[f]	12.2[g], 4.1[h], 3.2[i], 15.7[j]	
Mannosan TC	%	0.25 ± 0.09	0.02 ± 0.01	4.21 ± 0.59	0.89 ± 0.17	0.43 ± 0.19	1.12 ± 0.37	0.74 ± 0.21		0.45[e]	0.57[i]	
Sugar Alcohols TC⁻¹	%	0.03 ± 0.02	<0.001	0.05 ± 0.02	0.02 ± 0.01	0.01 ± 0.00	0.01 ± 0.00	0.25 ± 0.11				
Inositols (allo, myo, scyllo) TC	%	0.02 ± 0.02	<0.001	0.01 ± 0.00	<0.01	<0.01	<0.01	0.19 ± 0.10	0.023[o]	0.012[f]		
Arabitol TC⁻¹	%	<0.01	<0.0001	0.04 ± 0.02	0.02 ± 0.01	0.01 ± 0.01	0.01 ± 0.00	0.07 ± 0.02	1.18[o]	0.05[e], 0.13[f]	0.21[i]	
Resin acids TC⁻¹	%	10.42 ± 4.67	0.55 ± 0.15	30.40 ± 11.63	26.87 ± 11.31	9.21 ± 1.41	5.25 ± 4.00	1.82 ± 1.11				
Dehydroabietic acid TC⁻¹	%	10.38 ± 4.66	0.54 ± 0.15	28.15 ± 10.62	25.44 ± 10.81	9.11 ± 1.33	4.90 ± 3.71	1.70 ± 1.05		3.1[b], 0.3[c]	0.53[g], 3.37[h]	
Pimaric acids TC⁻¹	%	ND[a]	<0.01	1.12 ± 0.65	0.56 ± 0.28	0.01 ± 0.01	0.04 ± 0.04	0.02 ± 0.01		0.23[b], 0.003[c]	0.13[g],0.29[h]	
Abietic acids TC⁻¹	%	0.04 ± 0.03	0.01 ± 0.01	1.13 ± 0.45	0.88 ± 0.24	0.09 ± 0.08	0.31 ± 0.26	0.09 ± 0.05		0.002[c]	1.52[g],0.42[h]	
Lignin derivatives TC⁻¹	%	0.02 ± 0.01	<0.01	0.22 ± 0.05	0.08 ± 0.02	0.01 ± 0.00	0.18 ± 0.16	0.58 ± 0.23				
4-hydroxybenzoic acid TC⁻¹	%	0.01 ± 0.01	<0.01	0.03 ± 0.01	0.04 ± 0.01	0.01 ± 0.00	0.03 ± 0.02	0.13 ± 0.05		0.057[f]		
pyrogallol TC⁻¹	%	<0.01	<0.001	0.18 ± 0.05	0.04 ± 0.02	<0.001	0.15 ± 0.14	0.42 ± 0.17				
shikimic acid TC⁻¹	%	ND	<0.0001	<0.01	<0.001	<0.0001	<0.001	0.04 ± 0.02				
Sum of polar organics TC	%	15.06 ± 4.34	1.28 ± 0.37	42.66 ± 9.97	35.90 ± 13.32	19.35 ± 3.17	9.07 ± 4.36	5.42 ± 0.97				
Ratios												
Levoglucosan K⁺	µg µg⁻¹	6.0 ± 1.2	0.8 ± 0.2	6.0 ± 1.5	51.5 ± 19.0	8.6 ± 2.8	3.1 ± 1.5	1.5 ± 0.3	2.00[m]	8.8[b], 20.6[c]	17.8[g], 2.9[i], 75.7[j]	
Levoglucosan/mannosan	µg µg⁻¹	44.7 ± 19.2	53.7 ± 19.0	2.2 ± 0.9	10.8 ± 3.5	25.5 ± 7.3	9.6 ± 7.0	3.5 ± 0.8	53.5[o]	14.2[e]	16.7[g],5.6[i],7.8[j]	

[a] ND:<detection limit.
[b] Lee et al., 2005.
[c] Yan et al., 2008.
[d] Alves et al., 2010.
[e] Claeys et al., 2010.
[f] Graham et al., 2002.
[g] Fine et al., 2001, 2002, 2004.
[h] Hays et al., 2002.
[i] Schmidl et al., 2008a.
[j] Schmidl et al., 2008b.
[k] McDonald et al., 2000.
[l] McDonald et al., 2003.
[m] Zhang et al., 2010.
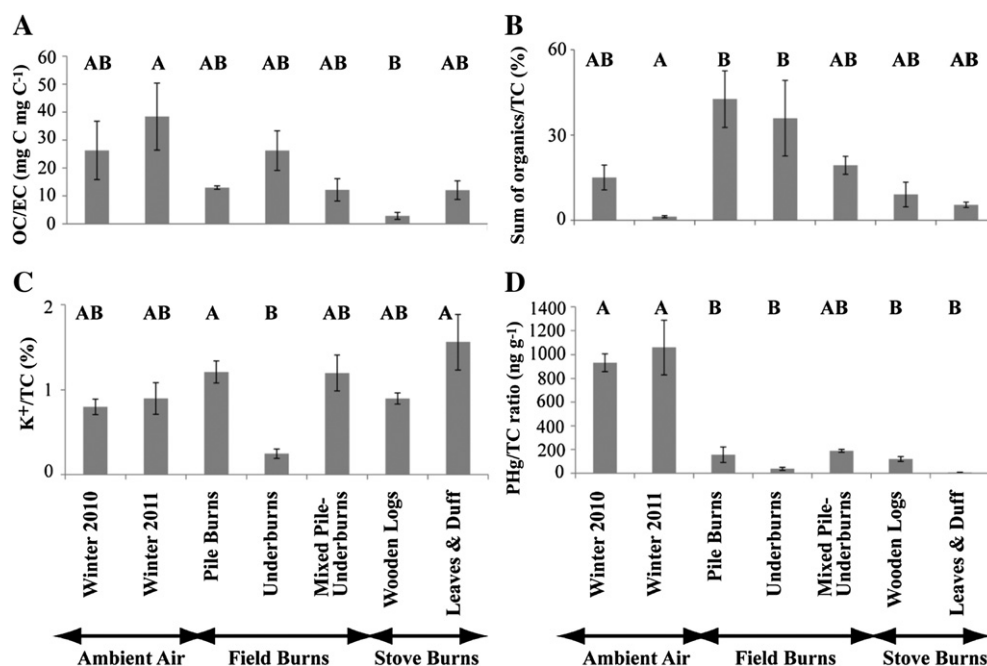[n] Watson and Chow 2001.
[o] Graham et al., 2003.

Fig. 1. Particulate-phase emissions summarized in groups, including ambient air of two respective years, field pile burns, underburns, and mixed pile-underburns, and stove burns using various tissues. (A) OC/EC ratios (in mg C mg^{-1} C); (B) Percentage of sum of polar organics to TC; (C) Percentage of K$^+$ to TC; (D) Ratio of particle-bound Hg (PHg) to TC. Bars labeled with identical letters within each figure panel are not statistically different from each other ($P > 0.05$). Statistical analyses showed overall statistically significant differences between groups in all panels; individual comparisons (Bonferroni post-hoc tests) indicate statistical differences when bars are marked with different letters (i.e., A is statistically different from B, but AB is not statistically different from A nor from B), although low sample numbers caused low statistical power.

sampling times, proximity to fires and smoke loads (e.g., ambient air versus prescribed fire plumes). For comparison of chemical composition, concentrations of compounds were hence normalized by total carbon (TC) concentration. Twelve polar organic compounds specific to biomass burning were analyzed in this study, and the sum of concentrations (standardized to TC) is shown in Fig. 1B and Table 1. In general, polar organics were emitted at a higher level from field burns than controlled stove burns, but we found no statistical differences between wooden tissues and green foliage. The level of organics in 2010 ambient air samples was much higher than that in 2011. We attribute the differences to more tourists visiting in 2010 when sampling occurred between Christmas and New Year and when snowy weather conditions may have caused high domestic biomass combustion activities; in 2011, we sampled prior to the peak tourist season before Christmas and sunny and warm weather may have led to lower domestic wood combustion.

Polar organic compounds were divided into four main groups: anhydrosugars, sugar alcohols, resin acids, and lignin derivatives. The fractional abundance of each group is shown in Fig. 2. Emissions in all types of burning were dominated by anhydrosugars (i.e., levoglucosan and mannosan, average of 48%) and resin acids (48%), followed by sugar alcohols (i.e., inositols and arabitol, 2%) and lignin derivatives (i.e., 4-hydroxybenzoic acid, pyrogallol, and shikimic acid, 2%). Inositols and arabitol only showed significant levels in stove burns of greenleaf manzanita, a common evergreen understory shrub of the basin. Levoglucosan, a sugar anhydride produced from cellulose during combustion, is widely reported as the major particulate organic compound emitted during biomass burning (Lee et al., 2005), and is the most commonly used organic tracer for biomass combustion (Simoneit et al., 1999). Previous studies showed that ratios of levoglucosan to OC varied among different plant tissues (i.e. branches > straw > needles > leaves; Sullivan et al., 2008). Mannosan also is an altered carbohydrate from biomass burning, with much lower abundance than levoglucosan

(Claeys et al., 2010). Levoglucosan and mannosan are expected to be abundant mainly in dry vegetation tissues such as wood, dry leaves, and dry pine needles (Schmidl et al., 2008a). However, we did not observe such patterns in our study: no differences in anhydrosugars were observed between fuel types, although we observed generally higher anhydrosugars levels in stove burns compared to prescribed burns. Percentages of levoglucosan to TC in emissions measured in our study are generally in the range of other studies (Table 1), except that the levoglucosan levels from stove burning of logs (2.5 ± 1.0%) were lower compared to other studies (a mean of 12.2%: Fine et al., 2001; Fine et al., 2002, 2004; a mean of 15.7%: Schmidl et al., 2008b). The ratio of levoglucosan to mannosan in this study also falls within the range of other studies (Table 1); however, levoglucosan to mannosan ratios in stove burns of logs (composed of cedar, fir and pine) in our study were lower than that from hardwood smoke in fireplace combustion (Fine et al., 2001, 2002, 2004); this could potentially indicate that the ratio of levoglucosan to mannosan is different for different wood types.

Sugar alcohols, including inositols (allo-inositol, myo-inositol and scyllo-inositol) and arabitol, are generally expected to be prevalent in green vegetation (Schmidl et al., 2008a). In support of this notion, the only significant contribution of inositols and arabitol in our study was observed in stove burning of green manzanita leaves and branches, and negligible levels were present in other burn types. No significant levels of inositols and arabitol, however, were found in field underburns either where manzanita foliage also was subject to burning; this is possibly due to relative low mass loss of manzanita foliage compared to mass loss of other tissues (including litter, and woody logs and branches).

Resin acids are prevalent in conifer tissues such as wood, bark and foliage (Hays et al., 2002; Medeiros and Simoneit, 2008; Simoneit et al., 1993), and studies showed them predominant in the burning of softwood (conifers) during wildland fires (Zielinska and Samburova, 2011). Similarly, Iinuma et al. (2007) showed that pine
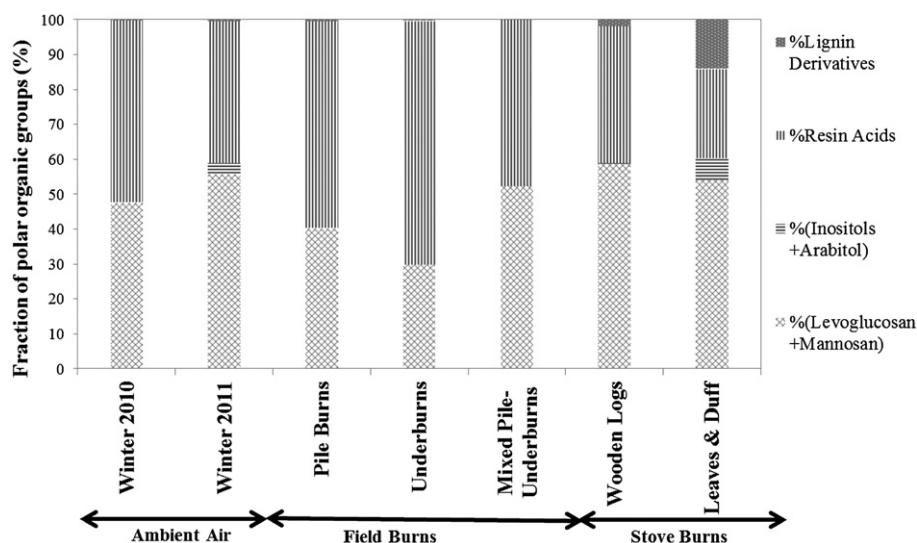
Fig. 2. Fractional representation of individual groups of polar organic compounds to sum of organics. The four classes include lignin derivatives, resin acids, inositols plus arabitol, and levoglucosan plus mannosan. Resin acids include the sum of pimaric acid, abietic acid, and dehydroabietic acid; lignin derivatives include 4-hydroxybenzoic acid, pyrogallol, and shikimic acid.

and pine with green needles had a higher percentage of resin acids than savanna grass. Abietic acid was used as a tracer to differentiate hardwood from softwood burning by Schmidl et al. (2008b). Pimaric acid and abietic acid are emitted by direct vaporization, while dehydroabietic acid has been regarded as a pyrolysis product of resin acids and proposed as a molecular tracer for coniferous wood combustion (Medeiros and Simoneit, 2008; Simoneit et al., 1993; Standley and Simoneit, 1994). In our study, resin acids are reported as the sum of pimaric acid, abietic acid, and dehydroabietic acid. For total resin acids, both field burns and stove burns showed higher levels in wooden tissues than in leaves and duff (Table 1), which supports the notion that resin acids may be a good tracer for softwood combustion. Resin acids, particularly dehydroabietic acids, show higher contributions in ambient air samples during the 2010 residential wood burning season than in 2011; this further indicates a higher abundance of wood combustion, specifically from coniferous species, in the 2010 sampling season. For abietic acid, previous studies showed higher contributions from leaf than wood burning (Schmidl et al., 2008a), but our study did not confirm this (no differences between wooden biomass and green foliage were observed).

Finally, lignin derivatives including 4-hydroxybenzoic acid, pyrogallol, and shikimic acid have been found to be present in green and dry vegetation (Medeiros and Simoneit, 2008). Although stove burning of leaves and duff showed higher lignin levels than others, the uncertainties and variability within various groups is too large to draw specific conclusions about their respective emissions in various biomass burning classes (Table 1).

3.3. Soluble potassium (K⁺)

Water-soluble potassium is widely used as an inorganic biomass burning tracer (McDonald et al., 2000; Schmidl et al., 2008a). Pio et al. (2008) found that soluble K^+ was possibly a better biomass burning tracer than levoglucosan during an intense forest fire. Some research, however, has also shown that soluble K^+ may not be an ideal biomass combustion tracer as it has other sources, including soil dust (Wang et al., 2005), marine salt (Duvall et al., 2008) and meat cooking (McDonald et al., 2003). In our study, soluble K^+ levels were in a narrow range and similar between biomass burning types (Table 1, Fig. 1C): highest emissions were

observed in stove burns of leaves and duff, followed by pile burns in the field, mixed pile burns/underburns, and stove burns of logs. Only one emission source, field underburns clearly deviated from the others with lower K^+ level. Observed K^+ levels in most burns, with the exception of field underburns, were higher than those observed in prescribed fires of pine trees in Georgia (Yan et al., 2008) and from prescribed burnings of a shrub-dominated forest with pine trees in Portugal (Alves et al., 2010). However, observed K^+ to TC ratios in our study agreed well with Lee et al. (2005) in biomass emissions from prescribed burning in pine-dominated forest areas in Georgia. Burn studies also showed ratios of 0.5% from softwood logs burned in fireplaces (McDonald et al., 2000), 0.2% from softwoods logs burned in a stove (Schmidl et al., 2008b), and 0.9% from both softwoods and hardwoods burned in a fireplace (Fine et al., 2001, 2002, 2004). Our controlled burn studies showed K^+ to TC ratios in wooden logs averaging $0.9 \pm 0.1\%$ and in leaves and duff of $1.6 \pm 0.3\%$. These results indicate that K^+ emissions are affected by biomass types, with leaves showing higher emission ratios compared to wood and with potential differences between different wood types. This is supported by the study of Schmidl et al. (2008a) who showed a contrast in ratios between burning of leaf tissues versus woody tissues and by McDonald et al. (2000) who reported that hardwoods burned in a fireplace emitted 3–4 times higher K^+ levels than softwoods.

The percentage of K^+ in TC in ambient air samples was in a similar range to that of most biomass burning emissions. These relatively high levels of K^+ in ambient air were surprising as we consider ambient air only partially influenced by biomass emission sources (see above). However, it is possible that ambient air is influenced by other sources as well, including meat cooking (Watson and Chow, 2001; McDonald et al., 2003) and possibly other sources such as soil dust (Duvall et al., 2008).

3.4. Ratios of levoglucosan to water-soluble K⁺

Both levoglucosan and water-soluble K^+ are common tracers of biomass burning, and several studies report their ratios (Table 1). Levoglucosan to K^+ ratios from stack burns of manzanita in a Fire Science Laboratory study in Missoula, MT (Sullivan et al., 2008) averaged 2.3, a value similar to those we observed in stove combustion of manzanita leaves and duff (1.5 ± 0.3) in our study

(Table 1). Ratios in stove burns of wooden logs (3.1 ± 1.5), however, were lower compared to fireplace combustion of woods reported in Fine et al. (a mean of 17.8; 2001; 2002; 2004) and Schmidl et al. (a mean of 75.7; 2008b). Similarly, ratios from pile burns (6.0 ± 1.5) in our study were lower compared to a prescribed fire plume of pine trees in Georgia (a mean of 20.6; Yan et al., 2008), although they were similar to pine-dominated prescribed burns in Georgia (a mean of 8.8; Lee et al., 2005) and a Montana wildfire (a mean of 9; Ward et al., 2006).

Levoglucosan to K^+ ratios showed a correlation to OC/EC ratios across all data points ($r^2 = 0.82$). However, this correlation was driven by three data points with particularly high OC/EC ratios (above 20) and corresponding high ratios of levoglucosan to K^+, while data points with OC/EC ratios below 20 did not show such a correlation. Schkolnik et al. (2005) and Gao et al. (2003) reported that smoldering combustion (with their high OC/EC ratios, see section 3.1) tended to have higher levoglucosan/soluble K^+ emissions ratios compared to flaming combustion. Gao et al. (2003) ascribed this to the effect of the combustion phase on the chemical composition of emitted particles. Flaming combustion with exothermic gas-phase reaction can oxidize organics to a greater extent, favoring re-polymerization of levoglucosan into polysaccharides (Kawamoto et al., 2003). Smoldering combustion with a slow solid-phase reaction process, on the other hand, produced more unbroken organics and reduced pyrolysis of levoglucosan, further enhancing the levoglucosan level in comparison to soluble potassium.

3.5. Particulate-bound Hg

Results from our study support the notion that significant amounts of PHg are emitted during wildland fires; for example, wildfires are recognized as significant natural sources of atmospheric mercury (Artaxo et al., 2000; Brunke et al., 2001; Friedli et al., 2001; Obrist et al., 2008; Sigler et al., 2003; Turetsky et al., 2006; Wiedinmyer and Friedli, 2007). PHg/TC ratios (Fig. 1D) of field burns and stove burns greatly overlapped; unexpectedly, however, lower PHg TC^{-1} emission ratios were observed in underburns in the field compared to pile burns and mixed pile/underburns, and similarly lower PHg TC^{-1} ratios were observed for leaves and duff combusted in the wood stove as compared to wooden logs. These patterns were unexpected for two reasons: first, green foliage and branches show much higher tissue Hg levels as compared to woody tissues (Obrist et al., 2011); this was confirmed in nearby Sierra Nevada forests (Obrist et al., 2009). Second, controlled laboratory combustion of biomass (Obrist et al., 2008) showed that a key factor for emissions of PHg is fuel moisture, with high moisture green fuels producing higher PHg emissions –up to 50% of total mercury emissions–compared to dry fuels. These patterns were not reflected in observed PHg TC^{-1} emission ratios in this study. It is possible that in the field, reasons for higher PHg emissions include higher combustion temperature during pile burns which might have mobilized additional Hg from the underlying soils that contains large pools of Hg (Obrist et al., 2012). Friedli et al. (2003) indicated that higher mercury emission factors in wildfires from temperate North American forests compared to laboratory combustion may have been due to additional mercury release from compacted organic matter and fire-heated soil. However, we do not know the reasons for lower PHg levels during green fuel combustion in the controlled stove burns.

We observed much higher PHg TC^{-1} ratios in ambient air as compared to levels observed in biomass smoke. The main reason for this is likely that ambient air has much lower C content (for example, averaging around 18% and 22% in January and December based on IMPROVE network data from Sierra Nevada; Hand et al.,
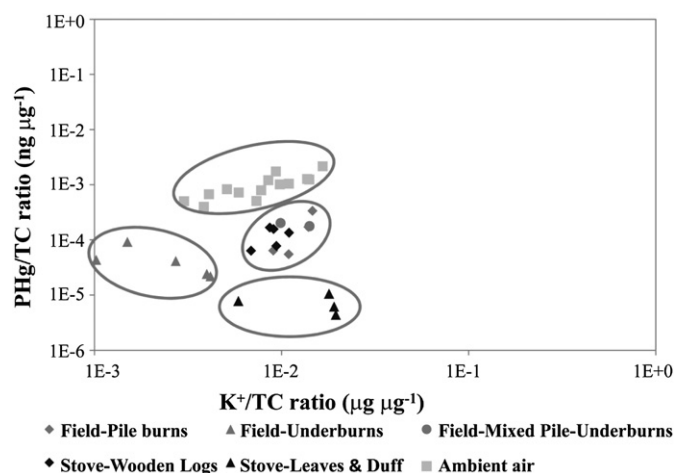

Fig. 3. Separation of samples based on a combination of PHg TC^{-1} and K^+ TC ratios.

2011) as compared to biomass smoke where most aerosols are mainly composed of carbonaceous material (>60% of PM mass; Chen et al., 2007). Hence, comparing PHg TC^{-1} ratios will ultimately show high ratios in ambient air. It is also possible that additional urban emission sources lead to higher PHg TC^{-1} ratios in ambient air, including from traffic (Landis et al., 2007).

3.6. Combined tracers to differentiate different types of biomass burning

A combination of multiple tracers may allow to separate different types of biomass burning. Using both PHg TC^{-1} and K^+ to TC ratios shows four clusters that are clearly separated as shown in Fig. 3. Ambient air shows highest PHg TC^{-1} ratios and is clearly separated from biomass burning emission types; at the same time, stove burning of green foliage with lowest PHg TC^{-1} ratios are separated as well. Adding K^+ to TC ratios separates prescribed field underburns from stove burns and field burns dominated by wooden biomass. However, separation of different burn types using multiple tracers shows limitations. First, the types of emissions samples (i.e., ambient air during wood combustion season, prescribed burning, and controlled stove burning) likely show regional characteristic, and results may not be applicable to other biomass burning sources outside of the Lake Tahoe basin. Secondly, we had limited sample numbers of each burning type, particularly with regard to controlled burns. However, the use of a combination of tracers allows separation of ambient air samples impacted by residential wood combustion from prescribed underburns and controlled stove burning of green foliage in the Lake Tahoe basin.

4. Conclusions

OC/EC showed higher ratios in green foliage combustion, indicating more moisture in green biomass tends to have the smoldering phase dominate during combustion. This is useful for differentiating between field pile burns and underburns, and stove burning of logs and leaves/duff. Anhydrosugars (levoglucosan/mannosan) and resin acids dominated among biomass burning groups and ambient air. A high percent of inositols and arabitol was observed in stove burning of green manzanita leaves. Levoglucosan showed higher levels in prescribed field burning than controlled stove burning, while resin acids had higher percentages in logs than green foliage both in field burns and stove burns. Nevertheless, large variations in abundance of molecular markers for biomass combustion between in-field prescribed burning and controlled

stove burns are due to the complex nature of combustion chemistry (Zielinska and Samburova, 2011). A combination of PHg TC^{-1} and K^+ to TC ratios may be utilized to differentiate the different biomass burning sources; in particular, we were able to distinguish ambient air (during residential wood combustion season) by their high PHg TC^{-1} ratios; emissions from green foliage combustion in stoves by their low PHg TC^{-1} ratios; and emissions from prescribed underburns by their low K^+ to TC ratio. Though not all burning types can easily be separated, this application along with indication from polar organic analysis on the burning environment and biomass types offered a way to characterize different biomass combustion types dominating in the Lake Tahoe basin.

Acknowledgments

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