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Emission of organic carbon, elemental carbon and water-soluble ions from crop straw burning under flaming and smoldering conditions

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ABSTRACT

Emissions from major agricultural residues were measured using a self-designed combustion system. Emission factors (EFs) of organic carbon (OC), elemental carbon (EC), and water-soluble ions (WSIs) (K^+, NH_4^+, Na^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}) in smoke from wheat and rice straw were measured under flaming and smoldering conditions. The OC_1/TC (total carbon) was highest (45.8% flaming, 57.7% smoldering) among carbon fractions. The mean EFs for OC (EF_{OC}) and EC (EF_{EC}) were 9.2 ± 3.9 and 2.2 ± 0.7 g/kg for wheat straw and 6.4 ± 1.9 and 1.1 ± 0.3 g/kg for rice straw under flaming conditions, while they were 40.8 ± 5.6 and 5.8 ± 1.0 g/kg and 37.6 ± 6.3 and 5.0 ± 1.4 g/kg under smoldering conditions, respectively. Higher EC ratios were observed in particulate matter (PM) mass under flaming conditions. The OC and EC for the two combustion patterns were significantly correlated ($p < 0.01$, $R = 0.95$ for wheat straw; $p < 0.01$, $R = 0.97$ for rice straw), and a higher positive correlation between OC_3 and EC was observed under both combustion conditions. WSIs emitted from flaming smoke were dominated by Cl^- and K^+, which contributed 3.4% and 2.4% of the PM mass for rice straw and 2.2% and 1.0% for wheat straw, respectively. The EFs of Cl^- and K^+ were 0.73 ± 0.16 and 0.51 ± 0.14 g/kg for wheat straw and 0.25 ± 0.15 and 0.12 ± 0.05 g/kg for rice straw under flaming conditions, while they were 0.42 ± 0.28 and 0.12 ± 0.06 g/kg and 0.30 ± 0.27 and 0.05 ± 0.03 g/kg under smoldering conditions, respectively. Na^+, Mg^{2+}, and NH_4^+ were vital components in PM, comprising from 0.8% (smoldering) to 3.1% (flaming) of the mass. Strong correlations of Cl^- with K^+, NH_4^+, and Na^+ ions were observed in rice straw and the calculated diagnostic ratios of OC/EC, K^+/Na^+ and Cl^-/Na^+ could be useful to distinguishing crop straw burning from other sources of atmospheric pollution.

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Introduction

Biomass burning is one of the major sources of atmospheric pollution, contributing many trace gases and aerosol pollutants to the regional and global atmospheric environment (Crounse et al., 2009; Marlier et al., 2013; Naeher et al., 2007). Many pollutants known to be hazardous to human health are released during burning of crop residues (Dennekamp & Abramson, 2011; Naeher et al., 2007; Price, Williamson, Henderson, Johnston, & Bowman, 2011; Torigoe et al., 2000), resulting in serious air pollution (Duan, Liu, Yu, & Cachier, 2004; Qin & Xie, 2011; Zhang et al., 2010). Pollution caused by burning in savanna areas (Andreae et al., 1996, 1998), the Amazon (Graham et al., 2002; Yamasoe, Artaxo, Miguel, & Allen, 2000), and wildfires (Alves, Gonçalves, Evtyugina et al., 2010; Alves, Gonçalves, Pio et al., 2010; Balasubramanian & See, 2006; Jaffe, Hafner, Chand, Westerling, & Spracklen, 2008; Liu, Kahn, Chaloulakou, & Koutrakis, 2009; McMeeking et al., 2006) has been extensively investigated. In southern Australia, biomass burning in rural communities was found to be dependent on season, fire activity, and duration of plume strikes (Reisen et al., 2011). Emissions from forest burning are the dominant contributors to total emissions among all land types in Southeast Asia (Shi & Yamaguchi, 2014). In China, approximately ~140 Tg of straw residue is burned annually (Cao, Zhang, Gong, & Zheng, 2008), accounting for about 30%–45% of the total energy consumption in rural areas (Zeng, Ma, & Ma, 2007).

Particulate matter (PM) emitted from crop residues burning contains a large fraction of carbonaceous and inorganic water-soluble ions (WSIs) (IPCC, 2013; Liousse et al., 1996; Pöschl, 2005). The two carbonaceous aerosol types are elemental carbon (EC), which is known to be an important contributor to radiative heating of the atmosphere, and organic carbon (OC), which is emitted along with EC, scatters radiation and has a cooling effect on the

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atmosphere (IPCC, 2001; Jacobson, 2001; Menon, Hansen, Nazarenko, & Luo, 2002). While high OC/EC ratios have been found in response to biomass burning, the warming potential brought by EC might be totally balanced by OC (Schaap & van der Gon, 2007). WSIs, which usually occupy one-third of the particulate matter mass, scatter incoming solar radiation to directly affect the balance of the Earth, on the other hand, by forming cloud condensation nuclei (CCN) with hygroscopic organic compounds to reduce solar radiation received on the surface of the Earth (Andrews et al., 2000; Chow et al., 2006; Penner, Dickinson, & O'Neill, 1992; Seinfeld & Pandis, 2012).

In previous studies, the relevance of organic and inorganic tracer species provides an overall characterization of the biomass burning smoke (Hays, Fine, Geron, Kleeman, & Gullett, 2005; Liu et al., 2014; Oanh, Albina, Ping, & Wang, 2005; Saarikoski et al., 2007; Simoneit et al., 1999; Ward, Hamilton, Dixon, Paulsen, & Simpson, 2006). The ratios of OC/EC in carbonaceous constituents have been shown to vary in diverse sources of combustion. Specifically, the OC/EC ratios range from 9.4–21.6 for biomass burning because of the high organic carbon content (Alves et al., 2011; Oros & Simoneit, 2001a, 2001b; Oros, bin Abas, Omar, Rahman, & Simoneit, 2006; Vicente et al., 2011), while the ratio of OC/EC in fossil fuel smoke is about 0.39 because of the higher EC concentrations (Wang, Lee, & Ho, 2006). In atmospheric aerosols, the OC/EC ratios range from 2.8 to 6.2 (Chen, Yin, Wei, & Yang, 2010; Tan et al., 2009). Among the WSIs particulate matter formed by biomass burning, K^+ is present in a relatively high concentration, and it has long been recognized as an important tracer of biomass burning in source apportionment studies (Chow, 1995). Potassium can also be used as an available tracer of biofuel combustion and biomass burning emissions (DeBell et al., 2004; Dibb et al., 1996; Duan et al., 2004; Hsu et al., 2009; Liu, Van Espen, Adams, Cafmeyer, & Maenhaut, 2000). The major ions emitted during burning of crop residues are K^+ and Cl^-, while those generated from forest fires are SO_4^{2-} and K^+ (Sillapapiromsuk, Chantara, Tengjaroenkul, Prasitwattanaseree, & Prapamontol, 2013). Furthermore, the diagnostic ratios of WSIs in fine particles can be used as reference indexes for characterization of the open burning of agricultural waste (Lin et al., 2012).

Field measured EFs of crop residues burning are crucial to emission inventory development; however, it is generally financially expensive and difficult to collect samples for such analyses. Although many laboratory simulations have investigated the emission characteristics of biomass burning (Cao et al., 2008; Frey et al., 2009; Oanh et al., 2011; Park, Sim, Bae, & Schauer, 2013; Zhang et al., 2007), few studies have investigated the composition of OC, EC, and WSIs emitted from crop residues under both flaming and smoldering conditions. Therefore, the main objectives of this study were to: (1) use a self-designed combustion device to simulate the open burning of agriculture residues; (2) measure the emissions of PM, OC, EC, and eight kinds of WSIs (Cl^-, NO_3^-, SO_4^{2-}, K^+, NH_4^+, Na^+, Mg^{2+}, Ca^{2+}) under flaming and smoldering conditions; (3) investigate the composition and diagnostic ratios of OC, EC, and WSIs; (4) characterize the properties of PM for straw burning and distinguish atmospheric pollution associated with burning of agricultural residues from other atmospheric pollution sources.

Experimental

Sample collection and pretreatment

Straw samples including six cultivars for rice and five cultivars for wheat were collected from different areas in China. Rice straw was collected from Zhejiang (cultivar: Nei2you6 and Yangliangyou6), Anhui (IIyou279 and Ningjing1), and Jiangsu (Hongliang166 and IIyou728). Wheat straw was sampled from Jiangsu (Yangmai10 and Yangmai16) and Henan (Aikang58, Wenliu, and Xinong979). Each straw sample consisted of leaves and stems. After removing the dust and debris, straw materials were air-dried indoors for at least 1 month, then cut into small pieces (about 6 cm long). The moisture content of each straw sample was measured before the experiment. Heavier moisture contents resulted in higher emission values. The EFs of $PM_{2.5}$ for rice straw were 18 and 5 g/kg, and the corresponding moisture contents were 25% and 10%, respectively (Carroll, Miller, Thompson, & Darley, 1977). On an air-dry basis, the moisture content for the wheat straw and rice straw were 13.2% and 12.2%. Before sampling, glass fiber filters were baked for 2 h at 500 °C, and then placed in desiccators for 24 h at room temperature to reach humidity equilibrium. After weighing, filter samples were wrapped in aluminum foil (baked at 500 °C for 2 h in advance) and preserved in a freezer at −20 °C.

Biomass burning simulation system

A combustion device was designed and installed in a 30 m^2 laboratory to simulate flaming and smoldering conditions (Fig. 1). The system consisted of four main parts, a combustion furnace, hood, U-shaped tunnel, and PM sampler (AH-200, Andersen, USA). The hood was cylindrical in shape, 0.48 m in diameter, and 1.12 m high, adopted a double-layer structure, was filled with cooling fluid in the interval. A U-shaped tunnel 3 m in length was immersed in the cooling tank. Both the hood and the U-shaped tunnel were made of stainless steel. Smoke emitted from the combustion furnace first went through the hood, where it cooled rapidly, then underwent secondary cooling in the U-shaped tunnel. Finally, the

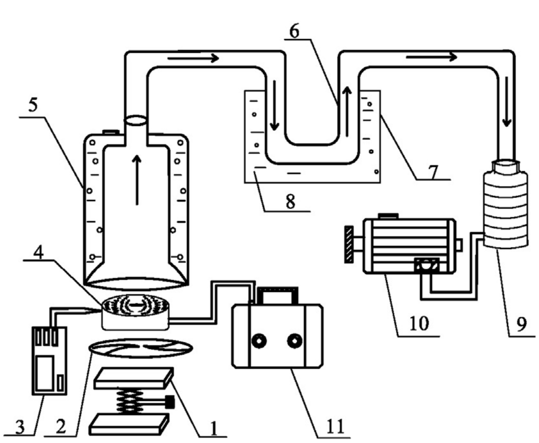

Fig 1. Schematic diagram of combustion device.

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