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Emission characteristics of carbonaceous particles and trace gases from open burning of crop residues in China

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HIGHLIGHTS

- Custom-made combustion chamber was used to determine emission factors (EFs).
- EFs of crop residue open burning specific to China and elsewhere were compared.
- Increased moisture content decreased CO_2 and enhanced CO, $PM_{2.5}$ & OC emissions.
- Emission inventories for crop residue combustion in China were compiled for 2008.

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ABSTRACT

Open burning of crop residue is an important source of carbonaceous pollutants, and has a large impact on the regional environment and global climate change. Laboratory burn tests were conducted using a custom-made combustion chamber to determine pollutants (i.e. CO_2, CO, $PM_{2.5}$, organic carbon (OC) and elemental carbon (EC)) emission factors (EFs) of wheat straw, rice straw and corn stalk; the three major agricultural crop residues in China. The average EFs were estimated to be 1351 ± 147 g kg^{-1} for CO_2, 52.0 ± 18.9 g kg^{-1} for CO, 10.6 ± 5.6 g kg^{-1} for $PM_{2.5}$, 4.8 ± 3.1 g kg^{-1} for OC and 0.24 ± 0.12 g kg^{-1} for EC. In addition, the effect of fuel moisture was investigated through the controlled burning of wheat straw. Increasing the moisture content decreased the CO_2 EF, and increased the EFs of CO, $PM_{2.5}$ and OC. Based on measurements from this study and nationwide statistics in crop type and area, pollutants emission inventories for crop residue combustion with $1^\circ \times 1^\circ$ resolution were compiled for 2008. Total emissions were 120 Tg CO_2, 4.6 Tg CO, 0.88 Tg $PM_{2.5}$, 0.39 Tg OC and 0.02 Tg EC.

1. Introduction

Open burning of crop residue is a common practice in China for the elimination of waste during the harvesting, post-harvesting or pre-planting periods. In Asia, field burning has become a serious concern due to its adverse environmental and health impacts (Bond et al., 2013; Fernandez et al., 2001; IPCC, 2013; Jacobson, 2001; Wei et al., 2015; Wu et al., 2013, 2012). Streets et al. (2003) estimated that open burning in China accounted for nearly half (~110 teragram [Tg]) of the total (250 Tg) crop residues burned in Asia in the mid-1990s. Huang et al. (2012a) reported a lower estimate of 40 Tg in 2006 due to the government's attempts to prohibit open burning in recent years. However, biomass burning emissions, including carbon dioxide (CO_2), carbon monoxide (CO), elemental carbon (EC), organic

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carbon (OC), particulate matter (PM) and others (Andreae and Merlet, 2001; Jenkins et al., 1992), still have significant impacts on the local and regional environment (Huang et al., 2014, 2012b).

Emission factors (EFs), defined as the mass of a pollutant emitted per unit of fuel consumed, are used to compile emission inventories, as inputs to dispersion models, and to evaluate the effectiveness of pollutant control strategies. EFs strongly depend on the type of crop, and burning conditions, such as fuel load and moisture content (Chen et al., 2010; McMeeking et al., 2009; Reid et al., 2005). Previous studies have obtained many EFs for open burning of crop residue worldwide as summarized in Supplemental Table S1 (e.g. Andreae and Merlet, 2001; Dhammapala et al., 2006; Hays et al., 2005; Kim Oanh et al., 2011; Nguyen et al., 1994; Turn et al., 1997; U.S. EPA, 1995; Yokelson et al., 2011), but few of these studies have considered the effect of moisture content on EFs (Kim Oanh et al., 2011). A more recent study by Hayashi et al. (2014) determined EFs for open burning of rice straw, wheat straw and barley straw in Japan using a portable combustion hood, and evaluated the effects of fuel moisture content on the EFs. Hayashi et al. (2014) found that an increased moisture content enhanced the emissions of CO, CH_4 and particulate organic matter. In China, few EFs from open burning of crop residue are available (Table S1). Li et al. (2007) reported EFs of $PM_{2.5}$ (particles with aerodynamic diameters <2.5 μm) and trace gases from open burning of wheat straw and maize stover in a rural area in Shandong Province. Zhang et al. (2008) performed burn experiments on rice, wheat and corn straw in a test chamber and measured EFs of trace gases. More recently, Zhang et al. (2013) conducted chamber burning experiments on rice straw and sugarcane leaves, the two major crop residues in Southeast China. EFs for both gaseous pollutants and particles were reported, including CO_2, CO, non-methane hydrocarbons, oxygenated volatile organic compounds, PM_{10} (particles with aerodynamic diameters <10 μm), $PM_{2.5}$, OC and EC. None of these studies considered the impacts of the fuel moisture content on pollutant emissions. Higher moisture often needs additional energy to vaporize the water and results in a lower combustion efficiency (CE) and higher pollutant emissions (Chen et al., 2010).

Given the limited availability of EFs in China, most emission inventories in China (e.g. Huang et al., 2012a; Streets et al., 2003; Yan et al., 2006) have used the EFs reported by Andreae and Merlet (2001) or Akagi et al. (2011). Biomass burning emissions are aggregated without specifying the fuel types or combustion conditions. This can cause large uncertainties when compiling emission inventories.

The objective of this study was to quantify EFs for gaseous and particle pollutants (i.e. CO_2, CO, $PM_{2.5}$, OC and EC) from major crop residues (i.e. wheat straw, rice straw and corn stalk) in China, using a laboratory combustion chamber. The impact of fuel moisture content on pollutant emissions was assessed because measured EFs are sensitive to moisture in the fuel. The results were compiled to produce an statewide emission inventory for 2008 with $1^\circ \times 1^\circ$ resolution.

2. Experimental section

2.1. Crop residue collection and processing

Wheat straw, rice straw and corn stalk were collected from five major crop producing regions (i.e. Shaanxi, Anhui, Shandong, Henan and Hebei Provinces). Samples were stored at ambient temperature (~20 °C) and humidity (35%–45%) for at least 1 month before the experiments. Ultimate analyses for the carbon (C) and nitrogen (N) content in dry mass, as well as proximate analyses for the moisture, ash, volatile matter, and fixed C content as received (Liao et al., 2004), were conducted (Table S2). When studying the effect of different moisture contents on emissions, we rehydrated the crop residues by adding ultrapure water to obtain fuels with different moisture levels (~10%, 28% and 50%), and then sealed the wet fuel in plastic bags for 1–2 days before combustion (Chen et al., 2010). The moisture content was tested before each burn.

2.2. Sampling and analysis

A combustion chamber was set up to simulate open burning of biomass at the Institute of Earth Environment, Chinese Academy of Sciences (IEECAS) in collaboration with the Desert Research Institute (DRI), USA. The chamber was a large cuboid container (1.8 (L) × 1.8 (W) × 2.2 m (H)) with a volume of approximately 8 m^3, with 3 mm-thick aluminum walls to withstand high temperatures. The combustion chamber was equipped with a thermocouple, a thermoanemometer, an air purification system and a sampling line to connect with a dilution sampler (Wang et al., 2012). The crop residues were first weighed with a balance (0.1–0.2 kg for each test) and then burned on a platform inside the combustion chamber. The smoke emitted from these laboratory burns was sampled by the dilution sampler and on-line instruments. The dilution ratios ranged from 5 to 15 in this study. The details of this biomass burning simulation system are described in Tian et al. (submitted manuscript, 2015). A total of 21 tests were conducted: nine for wheat straw, seven for rice straw, and five for corn stalk. The sampling periods typically lasted from 30 to 50 min.

$PM_{2.5}$ samples were collected from three parallel channels located downstream of the residence chamber of the dilution sampler, with a flow rate of 5 L min^{-1} per channel. Two 47 mm Whatman quartz microfiber filters (QM/A), which were pre-heated at 900 °C for 3 h before sampling to remove any residual carbon, were used for the carbon analysis, and one 47 mm Teflon-membrane filter (2 μm pore size, R2PJ047, Pall Life Sciences, Ann Arbor, MI, USA) was collected for gravimetric analyses. The sampled filters were stored in a refrigerator at ~4 °C before chemical analysis to minimize the evaporation of volatile components. Before and after sampling, the Teflon-membrane filters were conditioned for 24 h at ~25 °C and 35% relative humidity, and weighed using a microbalance with ±1 μg sensitivity (Sartorius, Göttingen, Germany). Each filter was weighed at least three times before and after sampling, and the net mass was obtained by subtracting the average of pre-sampling weights from the average of the post-sampling weights. The difference among the three repeated weights was less than 10 μg and 20 μg for a blank filter and a sampled filter, respectively. The OC, EC and their carbon fractions were analyzed following the IMPROVE_A thermal/optical protocol (Chow et al., 2007). Real-time CO levels and $PM_{2.5}$ mass concentrations were monitored by a CO analyzer (Model 48i, Thermo Scientific Inc., Franklin, MA, USA) and a DustTrak (Model 8532, TSI Inc., Shoreview, MW, USA) (Wang et al., 2009), respectively. Three nondispersive infrared (NDIR) CO_2 analyzers (Model SBA-4, PP System, Amesbury, MA, USA) were used to measure background CO_2, and CO_2 in stack and diluted emissions.

2.3. Determination of EFs

EFs were calculated by dividing the emission by the mass of the fuel consumed, and expressed as grams of emission per kilogram of consumed dry fuel ($g \cdot kg^{-1}$) (Andreae and Merlet, 2001). For particulate pollutants (i.e. $PM_{2.5}$, OC and EC), the EFs were calculated as:

$$EF_P = \frac{m_{filter}}{Q} \frac{V_{Total-chimney}}{m_{fuel}} DR \quad (1)$$

where EF_p is the EF of particulate pollutants for the specific crop