https://doi.org/10.5194/acp-2020-676
Preprint. Discussion started: 28 July 2020

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Chemical composition and light absorption of carbonaceous aerosols emitted from crop residue burning: Influence of combustion efficiency

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Abstract. Biomass burning is one of the major sources of carbonaceous aerosols, which affects air quality, radiation budget and human health. Field straw residue burning is a widespread type of biomass burning in Asia, while its emissions are poorly understood compared with the wood burning emissions. In this study, lab-controlled straw (wheat and corn) burning experiments were designed to investigate the emission factors and light absorption properties of different biomass burning organic aerosol (BBOA) fractions, including water soluble organic carbon (WSOC), humic-like substances (HULIS) and water insoluble organic carbon (WISOC). The influences of biofuel moisture content and combustion efficiency on emissions are comprehensively discussed. The emission factors of $PM_{2.5}$, OC and EC were 9.3±3.4, 4.6±1.9 and 0.21±0.07 g/kg for corn burning and 8.7±5.0, 3.9±2.8 and 0.22±0.05 g/kg for wheat burning, generally lower than wood or forest burning emissions. Though the mass contribution of WISOC among OC (32%-43%) was lower than WSOC, the light absorption contribution of WISOC (57%–84% @300-400 nm) surpassed WSOC due to the higher mass absorption efficiency (MAE) of WISOC. The results suggested that BBOA light absorption would be largely underestimated if only considering the water soluble fractions. However, the light absorption of WSOC among near-UV ranges, occupying 39%-43% of the total extracted OC absorption at 300 nm, cannot be negligible due to the sharper increase of absorption towards shorter wavelength compared with WISOC. HULIS were the major light absorption contributors among WSOC, due to the higher MAE of HULIS than other high-polarity WSOC components. The emission levels and light absorption of BBOA were largely influenced by the burning conditions, indicated by modified combustion efficiency (MCE) calculated by measured CO and CO_2 in this work. The emission factors of $PM_{2.5}$, OC, WSOC, HULIS and organic acids were enhanced under lower-MCE conditions or during higher-moisture straw burning experiments. Light absorption coefficients of BBOA at 365

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nm were also observed higher under lower-MCE conditions, which was mainly due to the elevated mass emission factors. Our results suggested that the influence of varied combustion efficiency on particle emissions could surpass the differences caused by different types of biofuels. Thus, the burning efficiency or conditions should be taken into consideration when estimating the influence of biomass burning. In addition, we observed that the K^+/OC and Cl^-/OC ratios increased under higher-MCE conditions due to the enhancement of released potassium and chlorine under higher fire temperatures during flaming combustion. This indicates that potassium ion, as a commonly used biomass burning tracer, may lead to estimation uncertainty if without considering the burning conditions.

1 Introduction

Biomass burning emissions, as a major primary source of carbonaceous aerosols, have significant effects on the air quality, human health as well as regional or global radiation budget (Bond, 2004; Chen et al., 2017a; Reid et al., 2005; Saleh et al., 2015). Biomass burning could contribute one-third of the black carbon (BC) budget and two-thirds of the primary organic aerosol budget on the global scale (Bond, 2004; Bond et al., 2013). In recent years, biomass burning organic aerosols (BBOA) also attracted much attention due to their substantial contribution to light-absorbing organic aerosols, known as brown carbon (BrC) (Andreae and Gelencsér, 2006; Laskin et al., 2015; Lin et al., 2016; Saleh et al., 2014; Washenfelder et al., 2015; Yan et al., 2018). Emission factors (EF) of BrC ranged from 1.0 to 1.4 g/kg biomass, comparable to those of BC (Aurell and Gullett, 2013). Majority of BrC aerosol mass was associated with biomass burning emissions in rural southeast US (Washenfelder et al., 2015). Regional radiative forcing effects of BrC could be comparable to those of BC over major areas dominated by biomass burning and biofuel combustion, such as South and East Asia (Feng et al., 2013).

Emission factors, chemical compositions and light absorption properties of biomass burning aerosols could be obviously influenced by different types of biomass, biofuel structures, moisture contents, and especially varied burning conditions (Chen and Bond, 2010; Holder et al., 2016; Reisen et al., 2018). The emissions of particulate organics could span several orders of magnitude depending on different burning conditions (Chen et al., 2017a; Jen et al., 2019). In general, higher levels of particulate matters (PM) and organic aerosols were emitted during less efficient biomass burning, due to prolonged incomplete or smoldering combustion (Holder et al., 2016; Jen et al., 2019; Reisen et al., 2018). Open biomass burning, especially smoldering combustion, dominates the organic carbon (OC) emissions in many regions of the world on an annual-average basis (Bond, 2004). The light absorption of BBOA are also largely dependent on the combustion conditions (Saleh et al., 2014). The contribution of BrC to aerosol light absorption at near-UV wavelength was reported higher for more smoldering combustion compared with more flaming combustion (Holder et al., 2016). The reported variation trends of BBOA absorption properties as a function of combustion conditions, however, are not consistent from

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different studies. High variability in reported emission factors and optical properties of BBOA from different burning conditions complicates their treatment in climate models (Liu et al., 2014; Saleh et al., 2014), and indicates the importance of further investigations on biomass burning emissions, especially the influence of burning conditions.

Unlike the well-understood BC, the light-absorbing OC or BrC comprise a wide range of poorly characterized organic compounds, which exhibit highly variable chemical and light absorption properties (Andreae and Gelencsér, 2006; Laskin et al., 2015; Lin et al., 2016; Saleh et al., 2014; Washenfelder et al., 2015; Yan et al., 2018). Previous studies have suggested that methanol extracted BrC were usually more light-absorbing than water extracts for BBOA or ambient aerosols (Chen and Bond, 2010; Liu et al., 2013). More than 92% of the light absorbing OC emitted from solid fuel pyrolysis could be extracted by methanol, compared with 73% for water-extracted compounds (Chen and Bond, 2010). Alkaline or methanol extracted OC fractions were also observed with higher mass absorption efficiency (MAE) at 365 nm than water soluble organic carbon (WSOC) for residential coal combustion (Li et al., 2018). Only considering the water soluble BrC would result in underestimation of BrC absorption and radiative forcing (Cheng et al., 2016; Cheng et al., 2017). Different light absorption properties of organic fractions could be attributed to the varied chemical compositions and structures (Chen et al., 2016a; Chen et al., 2016b; Chen et al., 2017b). However, few studies have been conducted to gain a comprehensive understanding on the influence of combustion conditions on the chemical composition and light absorption of different BBOA fractions.

Field open burning of agriculture wastes or crop residues is a widespread type of biomass burning in Asia (IARI, 2012; Bond, 2004; Streets et al., 2003b). Open crop residue burning during harvest season would result in severely adverse impacts on regional air quality and human health (Chen et al., 2017a; Li et al., 2014; Lin and Yu, 2011; Streets et al., 2003a; Zhang et al., 2010). The PM emission factors from agricultural waste burning range from 1.7 to 17.8 g/kg (Bond, 2004). Source apportionment results showed that ~50% of carbonaceous aerosols in Beijing were associated with biomass burning, with crop residue combustion as a major source (Cheng et al., 2013). Straw residue burning could contribute as high as 51% of PM and 76% of OC during harvest seasons in the agriculture regions in China (Li et al., 2014). Considering the large contribution of straw residue burning, the chemical compositions and light absorption properties of BBOA in Asia may differ from other regions with wood burning as the major type of biomass burning. However, the understanding on field straw residue burning emissions is still limited. A better characterization of the emission levels and optical properties of straw burning aerosols is required to quantify their effects on air quality and regional radiation forcing in agriculture area (Hungershoefer et al., 2008). Laboratory simulation experiment has been suggested as a good way to study biomass burning emissions due to its advantage in quantifying emission factors and controlling combustion conditions within well-defined limits. In this work, a series of lab burning experiments were designed to systematically investigate the emission factors, chemical compositions and light absorption properties of both water-soluble and water-insoluble carbonaceous aerosols

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emitted from straw residue burning. The influence of biofuel moisture contents, burning conditions and combustion efficiency on the BBOA emission levels and light absorption properties are comprehensively discussed.

2 Methods

2.1 Simulation and sampling of biomass burning aerosols

Lab-controlled burning experiments were conducted in the Laboratory of Biomass Burning Simulation at Peking University Shenzhen Graduate School. The simulation system was designed and optimized on the basis of the one used in He et al. (2010) (He et al., 2010), which included combustion system, dilution system, sampling system and data acquisition system (Figure S1). During each experiment, about 1-2 kg biomass fuels were ignited on the combustion pan. The emitted smoke was collected by the hood above the fire, and diluted by zero air (21 mol% O_2 and 79 mol % N_2) before collected on filters or monitored by online instruments. Smoke aerosols were collected on both Teflon (Whatman Inc.) and quartz fiber (Whatman Inc.) filters, using a $PM_{2.5}$ cutoff with a sampling flow rate of 16.7 L/min. During each burning experiment, CO and CO_2 were measured continuously by CO and CO_2 analyzers (Thermo Scientific Inc., Bremen, Germany). The burning efficiency, calculated based on the online CO and CO_2 data, were monitored continuously during each experiment (Table S1). The variation of fire temperatures during each experiment was also measured by a sensor above the fire (Figure S1).

In this study, corn and wheat, two primary grain crops in China, burning was simulated to represent the straw residue burning in China. To investigate the influence of biofuel moisture contents on burning emissions, straws with different levels of moisture contents were burned, including low (13%) and high (18%) levels for corn burning experiments, low (7%-9%), medium (18%-22%), and high (27%-33%) levels for wheat burning experiments (Table S1). The moisture content was measured by drying the biofuels in the oven at 105℃ for 24 h. Straw residues with different moisture contents were prepared by mixing weighted biofuels with weighted pure water in a plastic box, and shaking until the water was absorbed. Each experiment condition was repeated three times. All the conducted experiment conditions as well as burning conditions are summarized in Table S1.

2.2 Isolation of carbonaceous aerosols

The quartz fiber filters were used to extract different carbonaceous aerosol fractions, including water-insoluble organic carbon (WISOC), WSOC, and carbon component of HUmic-Like Substances ($HULIS_C$). The filter samples were firstly extracted in an ultrasonic bath twice using 10, and 10 mL ultrapure water, each time for 30 min. The extracts were then combined and filtered with a 0.45 μm pore size syringe filter (Gelman Sciences) to obtain the WSOC solutions. After removing the WSOC fraction on filters, the WISOC fractions were then extracted in an ultrasonic bath twice using 5, and 5 mL methanol, each time for 30 min. The extracts then were combined and filtered using a 0.25 μm syringe filter. The HULIS

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fraction was isolated from the WSOC solutions via solid phase extraction (SPE), with majority of low molecular weight organic acids (with relatively higher polarities) and sugars removed from the water solutions. Details about the HULIS extraction procedures were described in our previous paper (Wang et al., 2017). The WSOC fraction excluded HULIS was named as high-polarity WSOC (WSOC-h) in this study.

2.3 Quantification and light absorption measurements of carbonaceous aerosols

The total OC abundance was analyzed by a thermal/optical carbon analyzer (Sunset Laboratory). The concentrations of water soluble carbonaceous aerosol fractions, including WSOC and $HULIS_C$, were measured using a total organic carbon (TOC) analyzer (AnalytikJena multi N/C 3100). The WISOC concentrations were obtained by minus WSOC from the total OC. Light absorption of the extracted solutions (WSOC, $HULIS_C$ and WISOC) were measured by a UV-vis spectrometer (UV-1780, Shimadzu) over the wavelength range of 300-700 nm. The absorptions of WSOC and WISOC were added up to represent the absorption of the total extracted OC. The absorption coefficients (Abs_λ, Mm^{-1}) and mass absorption efficiency (MAE_λ) of isolated solutions at a wavelength λ were calculated as follow (Cheng et al., 2011; Cheng et al., 2016):

$$Abs_\lambda = (A_\lambda - A_{700})\frac{V_{\mathrm{sol}}}{V_{air} \times \mathrm{L}} \times \ln(10)$$

$$MAE_\lambda = \frac{Abs_\lambda}{C}$$

where A_λ and A_{700} represent the measured absorbance at wavelength λ and 700 nm. V_{sol} is the volume of extracted solutions and V_{air} is the volume of air sampled through the filter punch. The optical path length (L) is 1 cm in the present experiments. Ln (10) is used to convert from common logarithm to natural logarithm. C corresponds to the concentrations of OC, WISOC, WSOC or $HULIS_C$ fractions. It is noted that the total OC was used to represent the concentration of extracted OC, which may lead to an overestimation of WISOC mass concentration and an underestimation of MAE of WISOC. The wavelength dependence of light absorption is described using the Absorption Angstrom Exponent (AAE), which is calculated by a linear regression fit of $\log(Abs_\lambda)$ versus $\log(\lambda)$ in the wavelength range of 300-450 nm.

Water-soluble K^+, Cl^- and low molecular weight organic acids (acetic acid, formic acid, succinic acid, oxalic acid, propionic acid and methanesulfonic acid) were analyzed by ion chromatograph (DIONEX, ICS2500/ICS2000), following the procedures described in Guo et al. (2010) (Guo et al., 2010).

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3 Results and discussion

3.1 Burning conditions and combustion efficiency

The burning conditions and combustion efficiency, calculated by measured CO and CO_2 concentrations, of the simulation experiments are shown in Figure 1 and Table S1. Modified combustion efficiency (MCE), defined as $\Delta CO_2/(\Delta CO_2+\Delta CO)$, is used to indicate the burning conditions during a fire (Akagi et al., 2011; Andreae and Merlet, 2001). The burning conditions in this study varied from different fires, with the MCE ranging from 0.68 to 0.88 and an average value of 0.77. The amount and compositions of substances emitted from a given fire are determined to a large extent by the burning conditions or the ratio of flaming to smoldering combustion, which is often expressed as "combustion efficiency". Higher MCE (>0.9) indicates more flaming combustions, and lower MCE indicates more smoldering conditions. A previous study suggested that pure flaming has an MCE near 0.99, and the MCE of most smoldering combustion is around or lower than 0.8 (Akagi et al., 2011). The burning experiments were generally dominated by smoldering combustions in the present study.

The biomass fuels with lower moisture contents are generally burned more efficiently, with relatively higher MCE values (Table S1), which suggested higher proportion of flaming combustion during the fire. The MCE of higher-moisture biomass burning was generally lower, and prolonged smoldering combustion was observed (Figure 1, Table S1). Previous lab-controlled burning experiments also reported similar phenomenon that higher fuel moistures would lower the combustion efficiency, shorten flaming phase and introduce prolonged smoldering combustion (Chen et al., 2010). The relative proportion of flaming versus smoldering phases can vary considerably as a function of fuel moistures and structures (Andreae and Merlet, 2001).

Figure 1 displays variations of the monitored parameters (CO, CO_2, $\Delta CO/\Delta CO_2$ and fire temperature) during two selected burning experiments (low-moisture biomass burning with MCE=0.83, and high-moisture biomass burning with MCE=0.68). Different burning conditions dominate at different periods of a fire and the length of each period varied by experiments (Figure 1). Actually, flaming and smoldering phases occur simultaneously during a fire and the proportions of different combustion types vary over time (Akagi et al., 2011; Andreae and Merlet, 2001). For example, the initial period of low-moisture biomass burning experiment (Figure 1a) is dominated by flaming, wherein CO_2 increased rapidly to the highest level and $\Delta CO/\Delta CO_2$ ratios were lower (MCE was higher) compared with the smoldering-dominated period. The fire temperatures were very high during this initially high-efficiency burning period. During the later period, smoldering dominated the burning conditions. The burning efficiency and fire temperatures decreased during this period, and $\Delta CO/\Delta CO_2$ ratios were higher than the first period. Previous ground-based and aircraft measurements of wildfire emissions also observed gradually decreased combustion efficiency of a fire over time (Collier et al., 2016). For the high-moisture

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biomass burning, smoldering combustion dominated the fire types during the whole period (Figure 1b). Dehydration of the higher moistures from biofuels consumed more heat released from the combustion, thus the burning efficiency and fire temperatures were lower than those of the low-moisture biomass burning.

3.2 Emission factors of carbonaceous aerosols

The average emission factors of $PM_{2.5}$, OC and EC were 9.3±3.4, 4.6±1.9 and 0.21±0.07 g/kg for corn burning and 8.7±5.0, 3.9±2.8 and 0.22±0.05 g/kg for wheat burning (Figure 2). The particle EFs of corn burning were higher than wheat burning, which is likely due to their different biomass structures, resulting in different pyrolysis temperatures and efficiency (Zanatta et al., 2016). The measured emission factors in this study fall within the range of previous straw burning experiments (4.7-12.9, 1.2-8.9, 0.17-1.2 g/kg for $PM_{2.5}$, OC and EC, respectively)(Akagi et al., 2011; Hays et al., 2005; Li et al., 2007). The estimated EFs from crop residue burning were generally lower than wood or forest burning emissions (Akagi et al., 2011; Aurell and Gullett, 2013; Jen et al., 2019). However, open crop residue burning in the field could result in severe air pollution during harvest season, especially in agriculture areas in China and South Asia (IARI, 2012; Li et al., 2014; Streets et al., 2003b; Venkataraman et al., 2006). This type of biomass burning cannot be negligible in these regions.

Organic matter (OM), calculated by multiplying OC by 1.3 (Li et al., 2007), was the dominant component of straw burning aerosols, which accounted for ~64% and ~55% of the $PM_{2.5}$ emitted from corn and wheat burning (Figure 2). Around 57% and 68% of the OC from corn and wheat burning are water soluble, and $HULIS_C$ represent 53% and 46% of the WSOC. Though the mass contributions of WISOC were lower than WSOC in straw burning aerosols (Figure 2), the WISOC fractions cannot be negligible, especially for considering the light absorption properties of BBOA (see section 3.4). Previous studies also suggested a large portion of WISOC in ambient aerosols, which are important contributor of light-absorbing BrC (Cheng et al., 2016; Cheng et al., 2017).

The average EFs of water-soluble acetic acid, formic acid, succinic acid and oxalic acid were respectively 13.3±13.9, 4.1±3.3, 8.8±10.6, 2.2±1.1 mg/kg for corn burning and 13.0±14.5, 4.7±5.3, 9.9±13.5, 3.1±1.9 mg/kg for wheat burning (Figure 2). Propionic acid and methanesulfonic acid in most samples were below the instrument detection limits in this study, and their emissions were not taken into consideration in the following discussion. The quantified water-soluble low-molecular-weight acids averagely accounted for 0.84% (0.16%-1.6%) and 0.88% (0.24%-1.8%) of the water-soluble OM (WSOM) emitted from corn and wheat burning. Previous study has suggested that low molecular weight organic acids represented an important fraction of WSOC in BBOA, and oxalic acid was a dominated short dicarboxylic (C2-C6) acids (Falkovich et al., 2005). The estimated emission factors of acetic acid and formic acid in this work were lower than those emitted from eucalypt forest fires, which were reported 17 and 26 mg/kg for flaming combustion, and 104 and 94 mg/kg for smoldering combustion based on ground-based field measurements (Reisen et al., 2018). The difference could be attributed

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to different biofuels, burning conditions as well as conducted experimental methods.

Figure 2 compares the emission factors of $PM_{2.5}$, carbonaceous aerosols and low molecular weight organic acids from straw residue burning under different levels of moisture contents. The EFs of fine particles and organic carbonaceous aerosols from high-moisture biomass burning were obviously higher than those from low-moisture biomass burning. Substantial particulate carbonaceous aerosols could be generated from burning of higher-moisture biofuels, which is mainly associated with the prolonged smoldering phases and less efficient combustions (Figure 1, Table S1). Similar variation trends were also reported in previous biomass burning studies (Chen et al., 2010; Sanchis et al., 2014). Different levels of biofuel moisture contents will actually influence the burning conditions, and thus impact the emission levels and compositions of particulate matters.

3.3 Influence of combustion efficiency on emission factors

As shown in Figure 3, the emission factors of $PM_{2.5}$ and organic carbonaceous components increased with decreasing MCE. Particle emissions were obviously enhanced under less efficient burning conditions. The emission factors of $PM_{2.5}$, OC, WSOC and $HULIS_C$ from the most smoldering combustion experiment were about 3.4, 4.3, 3.8 and 2.8 times of those from the most flaming combustion condition, regardless of the biomass types. The emissions of low molecular weight organic acids also follow the similar variation trends with combustion efficiency as those of OC or WSOC emission factors (Figure S2). These trends are generally in agreement with previous studies (Dhammapala et al., 2006; Holder et al., 2016; Jen et al., 2019; Reisen et al., 2018; Wang et al., 2013). Under the same burning conditions, the emission factors of particles or organic aerosols from corn burning were slightly higher than those from wheat burning (Figure 3). This was mainly due to the different pyrolysis temperatures and combustion efficiency of different biofuels, which would influence the burning processes (Khan et al., 2009; Zanatta et al., 2016). Our results suggested that the influence of varied burning conditions or combustion efficiency on particle emissions could surpass the differences between the two types of straw residue burning measured in this study (Figure 3). Thus, the burning efficiency or conditions should be taken into consideration when simulate or estimate the influence of biomass burning emissions in future models.

Different from organic compounds, the emission factors of EC under different combustion efficiency remain relatively consistent (Figure 3e). Holder et al. (2016) summarized the results from lab and field studies, and also found that the black carbon emission factors from different studies are relatively constant, despite the differences in plume dilution or measurement methods (Holder et al., 2016). Some studies, however, reported an increasing trend in EC or BC emission levels with the increasing of combustion efficiency in wildfires or forest burns in U.S. (Aurell and Gullett, 2013; Jen et al., 2019). As the conducted experiments were mostly dominated by smoldering combustions (MCE=0.68-0.88) in this study, we cannot exclude the possibility that the EC emissions may be higher under flaming-dominated combustions (e.g. MCE>0.9).

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Though the less variations of EC emission factors as a function of MCE, a positive correlation between EC/(OC+EC) ratios and combustion efficiency was observed (Figure 3g). Due to the obvious dependence of EC/OC or EC/(OC+EC) ratios on burning efficiency, these ratios could be employed to indicate different burning conditions when the emitted CO and CO_2 data is not available, which have been used in previous studies (Xie et al., 2018; Xie et al., 2019).

To further investigate the influence of burning conditions on the chemical compositions of biomass burning aerosols, mass ratios of WSOC/OC, $HULIS_C$/OC, K^+/OC and Cl^-/OC as a function of burning efficiency are plotted in Figure 4. The WSOC/OC and $HULIS_C$/OC mass ratios ranged from 0.52-0.78 and 0.16-0.54 among different burning experiments. The $HULIS_C$/OC ratios were comparable to those (0.26-0.44, with an average of 0.34) reported in field or controlled chamber combustion experiments (Lin et al., 2010). We did not observe obvious variation trends of WSOC/OC or $HULIS_C$/OC ratios with MCE (Figure 4), which indicated relative constant BBOA chemical compositions under different combustion conditions. However, the K^+/OC and Cl^-/OC ratios showed consistent variation trends under different MCE conditions, which increased from <0.1 under the more smoldering condition to >0.5 under the more flaming condition for K^+/OC, and from 0.05 to >0.5 for Cl^-/OC (Figure 4). The highest K^+/OC (0.64) and Cl^-/OC (0.61) ratios were observed in a low-moisture wheat burning experiment with a MCE of 0.79. This is because that the K and Cl emissions from combustion are highly affected by fire temperatures and burning conditions. Lab-controlled experiments suggested that the proportions of released potassium and chlorine from the biomass fuels increase with the applied combustion temperatures (Jensen et al., 2000; Knudsen et al., 2004). The flaming combustion (with higher MCE) was observed much higher fire temperatures than the smoldering combustion (with lower MCE) (Figure 1). Though the emission levels of particles or organic aerosols decreased during higher efficiency burning (Figure 3), elevated proportions of potassium and chlorine were released into smokes during the flaming combustion phase under this condition (Figure 4b). Potassium ion is a commonly used tracer to indicate the biomass burning emissions. However, our results revealed that K^+ cannot correctly indicate the emission levels of biomass burning aerosols under obviously different burning conditions, which may lead to large uncertainty in estimating burning emissions if without considering the combustion conditions.

3.4 Light absorption of BBOA

The light absorption of straw burning organic aerosols decreased sharply from near-UV to visible wavelengths (Figure 5), indicating their properties as biomass burning-generated BrC. The absorption of WISOC, WSOC and $HULIS_C$ at 300 nm was as high as 4.5, 15.2 and 11.2 times of those at 400 nm for corn burning emissions, and 4.8, 9.2 and 10.6 times for wheat burning emissions. The wavelength dependence property of BBOA light absorption was described by AAE derived from the absorption in the range of 300-450 nm. The AAE of WISOC, WSOC and $HULIS_C$ were respectively 5.8-5.9, 8.6-11.3, 8.9-10.2 for corn burning aerosols and 5.7-6.0, 8.1-9.0, 9.0-10.5 for wheat burning aerosols, and the averaged values were

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also shown in Figure 5. The water-soluble BBOA fractions (WSOC and HULIS) showed stronger wavelength dependence than the water insoluble fractions. The estimated AAE values of straw burning organic aerosols in this study are comparable to those of BBOA (5.3-8.1) and biomass burning-influenced atmospheric aerosols (5.2-9.4) reported in previous studies (Hecobian et al., 2010; Hoffer et al., 2006; Wu et al., 2018; Wu et al., 2019; Xie et al., 2017; Xie et al., 2019; Zhu et al., 2018). The strong light absorption of biomass burning-generated BrC in near-UV range would lead to an increase in aerosol light absorption and radiative forcing efficiency (Chakrabarty et al., 2010).

The WISOC was the most important light-absorption fraction among straw burning organic aerosols, which contributed 61%–84% and 57%–72% of the light absorption (@300-400 nm) by extracted BrC emitted from corn and wheat burning (Figure 5). $HULIS_C$ and other high-polarity WSOC (WSOC-h=WSOC-$HULIS_C$) respectively occupy 16%-28% and 1%-10% of the BBOA absorption at 300-400 nm for corn burning, and 17%-29% and 12%-15% for wheat burning. Though the mass contribution of WISOC was lower than WSOC (Figure 2), the light absorption of WISOC surpassed WSOC due to the higher light absorption capability of water-insoluble BBOA, indicated by the higher MAE of WISOC (Figure 6). Meanwhile, the light absorption of water-soluble BBOA among near-UV ranges cannot be neglected due to their sharper increase of absorption towards shorter wavelength compared with WISOC (Figure 5). The light absorption contribution of WSOC to extracted BrC increased substantially from 16%-28% at 400 nm to 39%-43% at 300 nm. Among the water-soluble BBOA, HULIS were the major contributors of light absorption, which occupied 74% and 68% of the WSOC absorption at 300 nm for corn and wheat burning emissions, respectively. This was due to the higher light absorption capability of HULIS than other high-polarity WSOC fractions (Figure 6), though their mass contributions were comparable in straw burning aerosols (Figure 2).

The light absorption capabilities of different BBOA fractions are compared in Figure 6. The estimated MAE_{365} values of straw burning-generated BrC in this study are comparable to those reported in previous studies (Fan et al., 2018; Xie et al., 2017). The MAE of WISOC are higher than water-soluble BBOA (WSOC and HULIS) among the measured wavelength ranges for both corn and wheat burning aerosols. The MAE_{300} of WISOC was 1.6 and 1.7 times of WSOC emitted from corn and wheat burning, and comparable to those of HULIS (Figure 6). Due to the slower decrease of WISOC absorption towards visible wavelengths than the water-soluble fractions (Figure 5), the MAE_{365} of WISOC was as high as 2.5 and 2.2 times of WSOC from corn and wheat burning emissions, and 1.7 and 1.6 times of HULIS. Though the mass contribution of WISOC among BBOA could be smaller than WSOC, their contribution to light absorption cannot be neglected due to the higher MAE of water insoluble BBOA. The light absorption of BBOA would be largely underestimated if only considering the water soluble fractions. Previous studies also reported a large proportion of WISOC absorption in BBOA and ambient aerosols (Cheng et al., 2016; Cheng et al., 2017; Park et al., 2018; Sengupta et al., 2018).

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294 Figure 7 clearly shows the dependence of BBOA absorption coefficient (Abs_{365}) on burning conditions. Higher Abs_{365}
295 of biomass burning-generated BrC were observed under less efficient burning conditions for both corn and wheat burning
296 experiments. This is mainly due to the elevated BBOA emission factors as the decreasing of MCE (Figure 3). Furthermore,
297 the MAE_{365} of WSOC and HULIS emitted from straw burning were slightly higher under less efficient burning conditions
298 (Figures 7d, 7e). For the WISOC, however, we did not observe obvious dependence of MAE_{365} on the combustion efficiency
299 (Figure 7f). Previous lab and field studies suggested that the optical properties of biomass burning aerosols are more
300 dependent on burning conditions other than fuel types (Liu et al., 2014; Xie et al., 2017). The MAE_{365} of BBOA emitted
301 from flaming combustion were reported higher than those from smoldering combustion based on lab-controlled burning
302 experiments (Xie et al., 2019). Another lab experiment also suggested the dependence of MAE_{365} of methanol-extracted
303 BBOA on burning conditions, while the variation trends are different regarding different fuel types or sampling methods
304 among different experiments (Xie et al., 2017). It is noted that limited sample population was selected to conduct the light
305 absorption measurements in this study, and we did not observe a significant correlation between MAE and combustion
306 conditions. More lab experiments are required to address the influence of combustion efficiency on light absorption
307 capability of biomass burning-emitted carbonaceous aerosols.

308 ## 4 Conclusions

309 The emission factors of $PM_{2.5}$, OC and EC were 9.3, 4.6 and 0.21 g/kg for corn burning and 8.7, 3.9 and 0.22 g/kg for
310 wheat burning, generally lower than wood or forest burning emissions. Around 57% and 68% of the OC emitted from corn
311 and wheat burning are WSOC, among which $HULIS_C$ represent 53% and 46% of the WSOC mass concentrations. Though
312 the mass contribution of WISOC was lower than WSOC, the light absorption contribution of WISOC (57%–84% @300-400
313 nm) surpassed WSOC due to the higher MAE of WISOC. The BBOA light absorption would be largely underestimated if
314 only considering the water soluble fractions. Meanwhile, the light absorption of WSOC among near-UV ranges, occupying
315 39%-43% of extracted OC absorption at 300 nm, cannot be negligible due to their sharper increase of absorption towards
316 shorter wavelength compared with WISOC. HULIS were the major light absorption contributors among WSOC, and their
317 light absorption capability was higher than other high-polarity WSOC components.

318 The emission levels, compositions and light absorption of BBOA were largely influenced by the burning conditions and
319 biofuel moisture contents. The combustion conditions varied from different burning experiments, with the MCE ranging
320 from 0.68 to 0.88. The emission factors of $PM_{2.5}$ and organic carbonaceous aerosols were obviously enhanced under less
321 efficient burning conditions (lower MCE). The emission factors of $PM_{2.5}$, OC, WSOC and $HULIS_C$ from the most
322 smoldering combustion experiment were about 3.4, 4.3, 3.8 and 2.8 times of those from the most flaming combustion
323 condition, regardless of the biofuel types employed in this study. The emission factors of $PM_{2.5}$ and carbonaceous aerosols

https://doi.org/10.5194/acp-2020-676
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from high-moisture straw burning were obviously elevated compared with those from low-moisture straw burning experiments. This is mainly due to the prolonged smoldering and incomplete combustion period during high-moisture biomass burning.

The EC/(EC+OC) ratios showed a positive correlation with MCE, though EC emission factors remain relative constant under different combustion conditions. Thus, it is reasonable to employ EC/OC or EC/(EC+OC) ratios as an indicator of biomass burning conditions. The mass ratios of WSOC/OC or $HULIS_C$/OC did not display obvious variation trends under different combustion efficiency. However, the K^+/OC and Cl^-/OC ratios showed continuous increasing trends during higher efficiency burning, from <0.1 under the more smoldering condition to >0.5 under the more flaming condition for K^+/OC, and from 0.05 to >0.5 for Cl^-/OC ratios. This is mainly attributed to the elevated proportions of released potassium and chlorine from biofuels under the higher fire temperatures during flaming combustions. Our results indicate that potassium ion, as a commonly used biomass burning tracer, may lead to large uncertainty in estimating biomass burning emission levels if without considering the combustion conditions.

Higher absorption coefficient (Abs_{365}) of straw burning-generated BrC, including WSOC, HULIS and WISOC, were observed under less efficient burning conditions for both corn and wheat burning. This is mainly attributed to the higher BBOA emission factors as the decreasing of MCE. In addition, the MAE_{365} of WSOC and HULIS emitted from straw burning were slightly higher under low-MCE conditions. Our results suggested that the influence of varied combustion efficiency on the emission levels and light absorption of BBOA could surpass the differences between biofuel types. Thus, the burning efficiency or combustion conditions should be taken into consideration when estimate the influence of biomass burning.

Data availability. The data presented in this article are available from the authors upon request (minhu@pku.edu.cn).

The Supplement related to this article is available online

Author contributions. MH, ZW, XH, and LH organized the project. YW conducted the simulation experiments. YW, NX and YQ analyzed the samples. YW wrote the manuscript with input from all co-authors. All authors contributed to discussing the results and commenting on the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

https://doi.org/10.5194/acp-2020-676
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Acknowledgements. This work was supported by National Natural Science Foundation of China (91844301, 91544214), and the project funded by China Postdoctoral Science Foundation (2019M650354). We also thank Dr. Song Guo for his helpful suggestions on this work.

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Figures

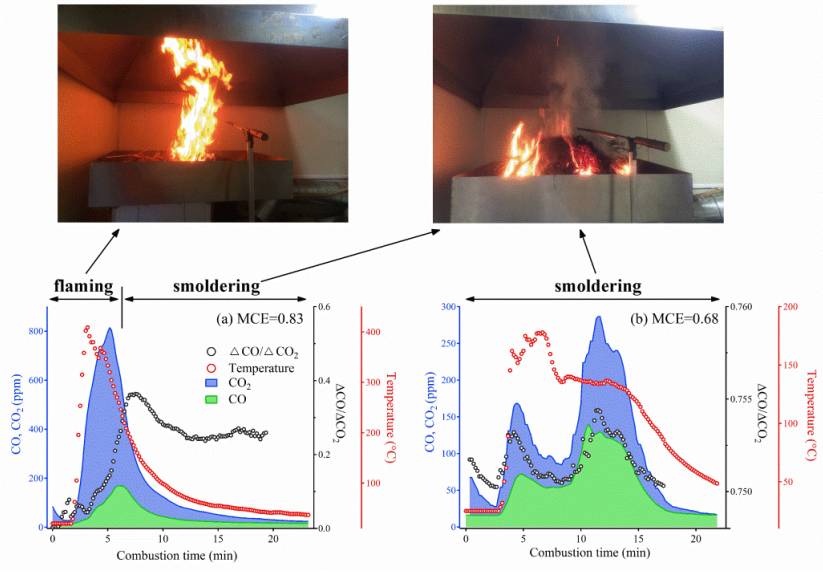

Figure 1 Variations of measured CO, CO_2 concentrations, $\Delta CO/\Delta CO_2$, fire temperatures and burning conditions during two selected experiments, with an averaged MCE value of (a) 0.83 and (b) 0.68.

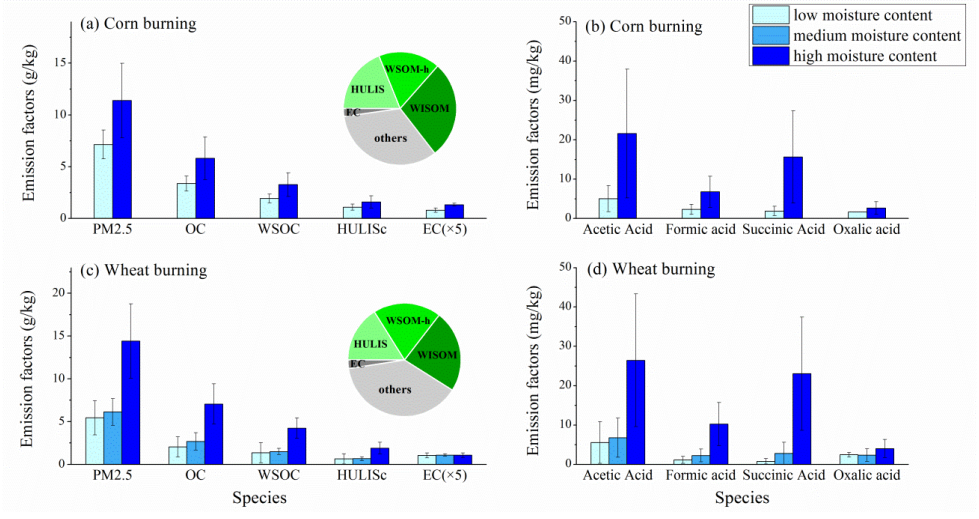

Figure 2 Emission factors of $PM_{2.5}$, OC, WSOC, $HULIS_C$ and EC from (a) corn burning and (c) wheat burning, and emission factors of low molecular weight organic acids (acetic acid, formic acid, succinic acid, and oxalic acid) from (b) corn burning and (d) wheat burning. The EC emission factors are represented by 5 ×EC due to the low values. The pie charts in panels (a) and (c) represent the contribution of major carbonaceous aerosols among $PM_{2.5}$. The high-polarity WSOM (WSOM-h) is calculated by subtracting HULIS from WSOM. Different moisture content levels correspond to those shown in Table S1.

https://doi.org/10.5194/acp-2020-676
Preprint. Discussion started: 28 July 2020

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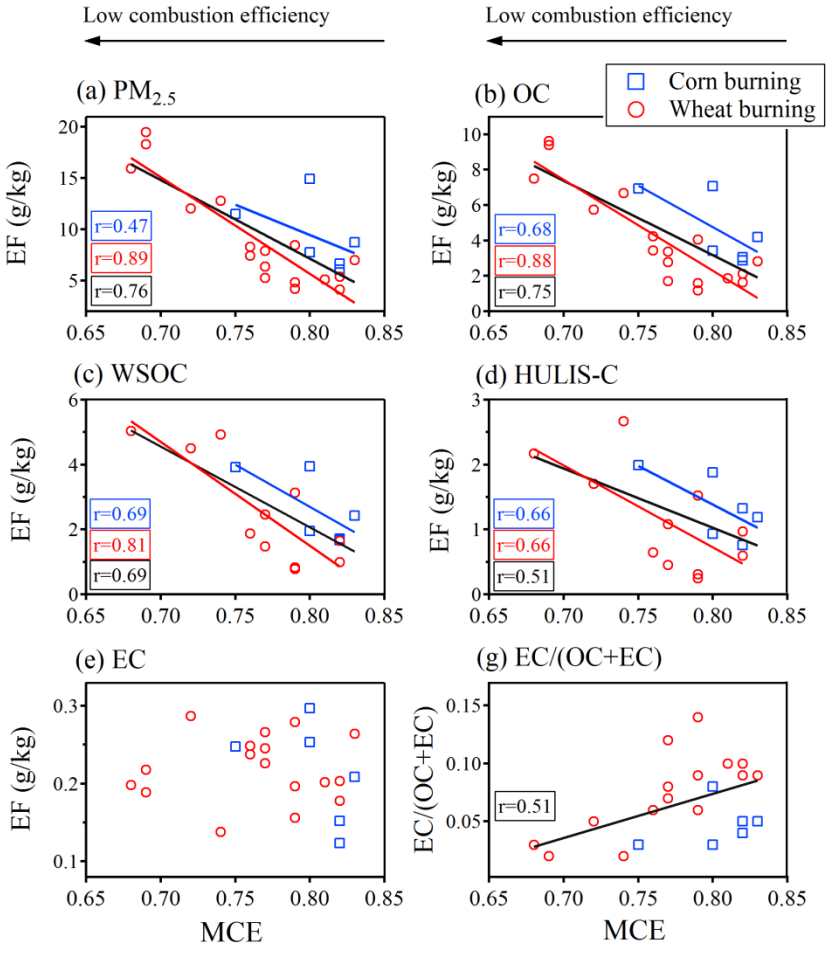

Figure 3 Emission factors of $PM_{2.5}$, carbonaceous aerosols (OC, WSOC, $HULIS_C$ and EC) and EC/(OC+EC) ratios as a function of modified combustion efficiency (MCE). Corn and wheat burning emissions are denoted by red and blue colors, respectively. The r values in each panel are the correlation coefficients between emission factors and MCE for corn (blue), wheat (red) and the overall (black) burning experiments.

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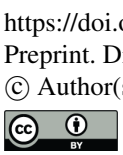

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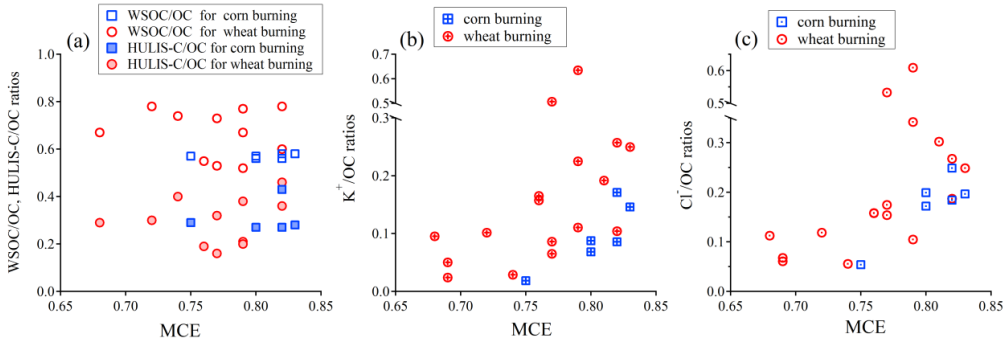

Figure 4 Variations of (a) WSOC/OC and $HULIS_C$/OC ratios, (b) K^+/OC, and (c) Cl^-/OC ratios as a function of modified combustion efficiency (MCE) for corn and wheat burning experiments. Corn and wheat burning emissions are denoted by red and blue colors, respectively.

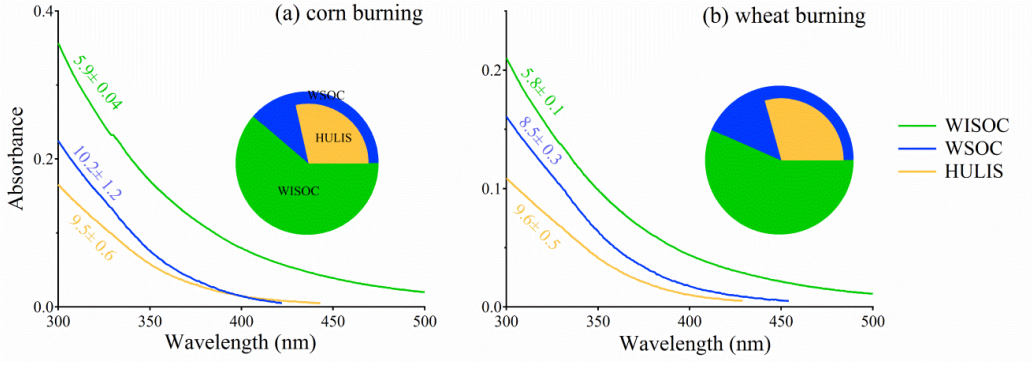

Figure 5 UV-vis spectra of carbonaceous aerosol solutions, including WSOC, $HULIS_C$ and WISOC, from (a) corn and (b) wheat burning experiments. The pie chart in each panel is the absorption contribution of different BBOA fractions at 300 nm. The number represents the average AAE of each BBOA fraction derived from the absorption in the wavelength range of 300-450 nm.

https://doi.org/10.5194/acp-2020-676
Preprint. Discussion started: 28 July 2020

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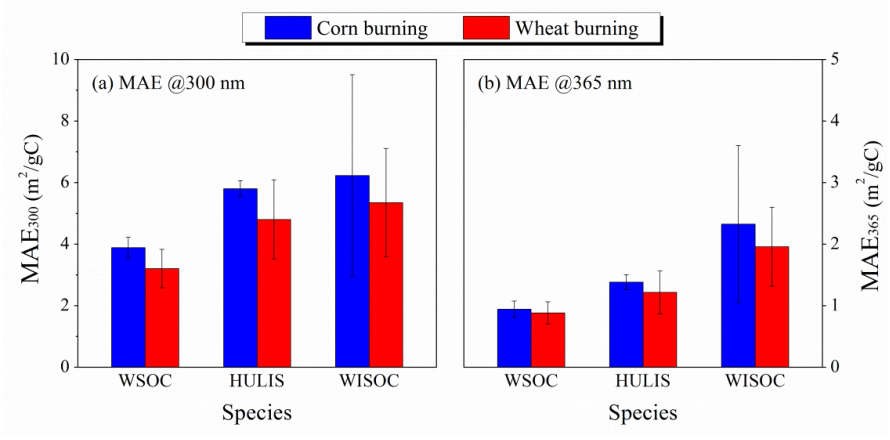

Figure 6 Mass absorption efficiency (MAE) of different organic carbonaceous aerosols, including WSOC, HULIS and WISOC emitted from corn and wheat burning.

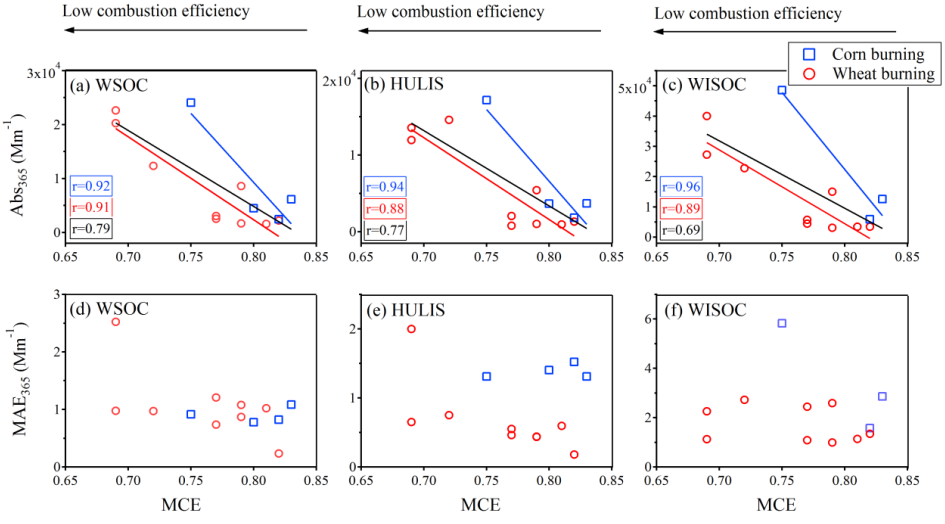

Figure 7 (a-c) Light absorption coefficients (Abs_{365}) and (d-f) mass absorption efficiency (MAE_{365}) of WSOC, $HULIS_C$ and WISOC at 365 nm as a function of combustion efficiency. Corn and wheat burning emissions are denoted by red and blue colors, respectively. The r values in panels (a-c) are the correlation coefficients for corn (blue), wheat (red) and overall (black) burning experiments.