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Emission factors of particulate matter and elemental carbon for crop residues and coals burned in typical household stoves in China

Guofeng Shen¹, Yifeng Yang¹, Wei Wang¹, Shu Tao^{1,*}, Chen Zhu¹, Yujia Min¹, Miao Xue¹, Junnan Ding¹, Bin Wang¹, Rong Wang¹, Huizhong Shen¹, Wei Li¹, Xilong Wang¹, and Armistead G. Russell²

¹Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China

²School of Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta, Georgia, 30332, USA

Abstract

Both particulate matter (PM) and black carbon (BC) impact climate change and human health. Uncertainties in emission inventories of PM and BC are partially due to large variation of measured emission factors (EFs) and lack of EFs from developing countries. Although there is a debate whether thermal-optically measured elemental carbon (EC) may be referred to as BC, EC are often treated as the same mass of BC. In this study, EFs of PM (EF_{PM}) and EC (EF_{FC}) for 9 crop residues and 5 coals were measured in actual rural cooking and coal stoves using the carbon mass balance method. The dependence of the EFs on fuel properties and combustion conditions were investigated. It was found that the mean EF_{PM} were 8.19 ± 4.27 and 3.17 ± 4.67 g/kg and the mean EF_{EC} were 1.38 ± 0.70 and 0.23 ± 0.36 g/kg for crop residues and coals, respectively. PM with size less than 10 µm (PM10) from crop residues were dominated by particles of aerodynamic size ranging from 0.7 to 2.1 μ m, while the most abundant size ranges of PM₁₀ from coals were either from 0.7 to 2.1 μ m or less than 0.7 μ m. Of various fuel properties and combustion conditions tested, fuel moisture and modified combustion efficiency (MCE) were the most critical factors affecting EFPM and EFEC for crop residues. For coal combustion, EFPM were primarily affected by MCE and volatile matter, while EFEC were significantly influenced by ash content, volatile matter, heat value, and MCE. It was also found that EC emissions were significantly correlated with emissions of PM with size less than $0.4 \,\mu m$.

Introduction

Exposure to particulate matter (PM), especially fine PM is associated with a wide range of diseases, including respiratory infection, lung cancer, and bronchus¹. Smoke from household fuel combustion is a large risk factor for people, especially females in developing countries². PM also impacts on global climate change and it was suggested that the Asian Brown Cloud was partly due to cook stove emission³. As an important component of PM, black carbon (BC) is the third largest warming agent, following CO₂ and methane⁴. PM can be produced from direct combustion or atmospheric formation, while BC is mainly from combustion of carbon-based fuels⁵. It is believed that BC is dominated by elemental graphitic carbon⁶. Till

^{*}Corresponding author phone and fax: 0086-10-62751938, taos@urban.pku.edu.cn.

Supporting Information **available**: The following materials, including design of the stoves, fuel properties, combustion conditions, the measured EFs, size distributions of PM_{10} , the stepwise regression analysis, and the relationship between EF_{EC} and $EF_{PM0.4}$, are available free of charge via the Internet at http://pubs.acs.org.

For emission inventory development, statistics such as medians or means for emission factors (EFs) from the literature were usually adopted. Since the measured EFs often varied over orders of magnitude, variation in EFs was the primary source of the overall uncertainty of emission inventories^{5, 10-11}. In addition, most reported EFs were measured in developed countries potentially biasing the global EF databank and likely leading to considerable underestimation of emission in developing countries^{5, 10-12}.

year, of which 19.4 Tg/year was from domestic biomass combustion⁹. BC emission was

2.54 Tg/year in Asian in 2000^{10} and 8.0 Tg/year globally in 1996⁵.

Combustion of solid fuels, including crop residues and coals, in cooking stoves is among the most important sources of PM and BC, particularly in developing countries^{5, 10-11}. It was estimated that the total quantities of crop residue combusted in field and indoor in China were 151 and 333 Tg in 2003, respectively¹². EFs of solid fuel varied widely due to variations in fuel types, fuel properties, and burning conditions, leading to large uncertainties in emission inventory^{5, 10-11}. These tests were conducted either in stoves¹³⁻¹⁷, chambers¹⁸⁻²¹, or open-field²¹⁻²³. It has been shown that emissions from cooking stoves, combustion chambers, or in open-field could be very different due to the differences in oxygen supply and circulation^{7, 21, 22-25}. In addition, large difference in EFs among different kinds of solid fuels has been well documented^{13-14, 17}. To improve process understanding and reduce uncertainty in emission estimation, it is necessary to quantify the influences of fuel properties and combustion conditions on the emission.

With the largest population and economy, China consumes the largest portion of energy among all developing countries. The main objectives of this study were to measure and characterize PM and EC emissions from combustion of commonly used crop residues and coals in traditional stoves in China and to quantitatively evaluate the key factors affecting the emissions so as to have a better understanding of the variations of EFs of PM (EF_{PM}) and EC (EF_{EC}). The information provided is useful for improving emission inventories for PM and EC. Particle size distribution of the PM emission was also addressed since it is important in terms of environmental and human health impacts¹.

Methodology

Stoves and Fuels

Two types of stoves widely used in rural China are brick wok stoves designed for large-size round-bottom woks and movable cast-iron stoves for small woks, tea pot or other cookware. Both of them are fire enclosed and have been used for centuries²⁶. During a period from 1980s to 1990s, a campaign of disseminating fuel-saving stoves in rural China have been undertaken to improve fuel efficiency of these stoves by using taller chimney, smaller firebox and fire door, and shorter distance between grid and cookware. It was estimated that the total numbers of the improved brick and cast-iron stoves used in China were 143 and 349 million in 2006, respectively²⁶. In northern China, wok stoves used by 175 million rural residences are connected to heating beds, known as "Kang"²⁷. In this study, a brick wok stove for crop residue burning was set up in a rural kitchen and a cast-iron stove for coal combustion was purchased from the local market in suburban Beijing. The exited smoke

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from the wok stove (passed a "Kang") and cast-iron stove entered a mixing chamber (4.5 m³) with a build-in mixing fan. No further dilution was performed to avoid alterations in particulate mass loading and size distribution²⁴. The photos of the stoves are shown in the Supporting Information (S1). Nine crop residues which contributed more than 90% of the total crop residue combusted in China (rice 17.5%, wheat 19.5%, corn 39.1%, beans including soybean and horsebean 5.4%, cotton 2.4%, oil crop including peanut, sesame, and rape 9.3%)²⁸ and five coals (two honeycomb briquettes from Beijing and Taiyuan and three chunk coals from Taiyuan and Yulin) were tested. The two honeycomb briquettes (15 cm diameter and 11 cm thick with 16 holes) were made of either anthracite with 4% volatile matter (VM) (from Beijing) or low volatile bituminous with 15% VM (from Taiyuan). The three raw chunk coals (from Taiyuan and Yulin) were all medium volatile bituminous (MVB) with VM between 23 and 29 %. The fuel properties are listed in the Supporting Information (S2).

Combustion Experiments

Combustion experiments were conducted following traditional methods used by rural residents. For coal combustion, coal (ca. 800-1000 g) in the stove was first ignited outdoor using small wood chips. After the coal was ignited, the stove was moved into the kitchen and set up under a stainless hood. For crop residue burning, pre-weighed fuel (ca. 500-700 g) was inserted into the stove chamber in 8-10 batches, and the burning lasted for 20-30 minutes. The ash was collected, weighed, and analyzed for carbon content. The combustion experiment for each type of fuel was conducted twice as duplicates. CO₂ and CO were measured every 2 seconds with an on-line detector equipped with non-dispersive infrared sensor. The equipment was calibrated before each experiment. Exact duration, fire temperature, smoke temperature, and relative humidity of smoke were recorded during the combustion (No significant difference among crop residues or among coals were observed). Emissions of PM and EC vary over the whole burning period of crop residues, which can be at least divided into flaming (with obvious fire) and smoldering (without observed fire) phases. Both CO and CO2 increased in the flaming phase and decreased during the smoldering phase²⁴. The difference in PM and EC emissions between the two phases was expected. Therefore, in addition to the whole burning cycle experiment, the two phases were tested individually in duplicates for all crop residues.

Sample Collection and Measurement

Low-volume pumps (XQC-15E, Tianyue, China) with quartz fiber filters were used to collect PM (as total suspended particles) in the mixing chamber at a flow rate of 1.5 L/min. A nine stage cascade impactor (FA-3, Kangjie, China) with glass fiber filters was used to collect PM₁₀ samples with different aerodynamic diameter (D_a) (< 0.4, 0.4 - 0.7, 0.7 - 1.1, 1.1 - 2.1, 2.1 -3.3, 3.3 - 4.7, 4.7 - 5.8, 5.8 - 9.0, and 9.0 -10.0 µm) at a flow rate of 28.3 L/min. The filters were baked at 450 °C for 6 hours and stored in a desiccator for 24 hours prior to weighing and sampling. After sampling, particle-loaded filters were packed with aluminum foil and stored in a desiccator before further analysis. Gravimetric measurements were conducted using a high precision (0.00001g) digital balance. EC and organic carbon (OC) were analyzed using Sunset EC/OC analyzer (Sunset Lab, USA). Samples were also collected in the mixing chamber before the combustion experiment and measured for PM, EC, OC, CO, and CO₂ using the same methods. The results were used as procedure blanks and subtracted from those measured during combustion.

Data analysis

EFs of CO₂, CO, PM, EC, and OC were calculated using the carbon mass balance method¹³. As most of the released gaseous carbon was in the forms of CO₂ or CO, the total hydrocarbon species in the gaseous phase were neglected which may lead to an error of less

than 4%²⁴. Several parameters including modified combustion efficiency (MCE), products of incomplete combustion (PIC), burning rate (R_b), and carbon release rate (R_c) were calculated to quantitatively describe the combustion conditions. The non-parametric Wilcoxon test for paired samples and Spearman correlation analysis were applied for data analysis using Statistica at a significant level of 0.05. To characterize the effects of various factors including fuel properties and combustion conditions, stepwise regression was applied. Calculation of the carbon balance method and various combustion conditions are provided in the Support Information (S3 and S4).

Results

EF_{PM} of Crop Residues and Coals

The measured EFs of PM, EC, OC, CO₂, and CO for crop residues and coals are presented in the Supporting Information (S5). In brief, the means and standard deviations of EFs of CO₂ (1380 ± 120 g/kg) and CO (126 ± 47 g/kg) for crop residue burning were comparable with the published data¹³⁻¹⁴. EFs of CO₂ and CO for coal combustions varied between 291 and 2286 g/kg and between 35 and 288 g/kg, respectively, which also agreed with those previously reported¹³⁻¹⁴.

 EF_{PM} for crop residues varied from 3.41 ± 0.11 (cotton) to 16.8 ± 4.81 g/kg (rape) with a mean and a standard deviation of 8.19 ± 4.27 g/kg. Andreae and Merlet reviewed published EF_{PM} data and came up with a similar value of 9.4 ± 6.0 g/kg for domestic biomass combustion⁹. The measured EF_{PM} for coal combustion varied widely from 0.065 ± 0.002 for anthracite (honeycomb coals from Beijing) to 10.8 ± 0.55 g/kg for bituminous coals (raw chunk from Yulin) with a mean and a standard deviation of 3.17 ± 4.67 g/kg, depending on origin and type of the coals. Zhang *et al.* reported similar EF_{PM} of 8.05 (1.12 - 29.0) and 1.30 (0.026 - 10.0) g/kg for crop residues and coals burned in stoves, respectively¹³. The differences in EF_{PM} between anthracite and bituminous coals and between raw chunk and honeycomb coals are well recognized, and our results agreed well with those reported previously⁷, ²⁹.

For the two phases of flaming and smoldering burning, the EFs of CO₂ and CO for crop residues were not significantly different (p > 0.05), while EF_{PM} of flaming phase (9.51 ± 3.02 g/kg) were significantly higher than those of smoldering one (7.09 ± 3.87 g/kg) (p < 0.05). In fact, smoke observed in smoldering phase was less thick than that in flaming phase. Although it was reported that PM number measured in flue gas during smoldering phase of biomass burning was lower than that of flaming phase, the fuel consumption rate was not measured for the two phases and the difference in EF_{PM} between the two phases were not calculated by them^{15, 18}. Taking high variability in burning conditions into consideration, the difference and the reasons causing such a difference should be further investigated.

EF_{EC} for Crop Residues and Coals

 EF_{EC} and EF_{OC} for crop residue burning were 1.38 ± 0.70 and 1.45 ± 0.62 g/kg, respectively. It is interesting to compare our results with those reported by the others for a better understanding of wide variation of EF measurements. In general, our results are more or less similar to those measured using cooking stoves in the literature^{25, 30}. For example, Li *et al.* reported that EF_{EC} and EF_{OC} for crop residues in residential stoves were 0.09 - 0.94and 0.85 - 3.21 g/kg, respectively²⁵. It is noted that the EF_{EC} , but not EF_{OC} , measured for residential stoves (both our study and those reported in the literature) were often higher than those measured in laboratory chambers or open field. It was reported that EF_{EC} and EF_{OC} were 0.08 and 6.2 g/kg for rice residue²⁰ and 0.35 and 1.9 g/kg for wheat²³ burned in laboratory chambers. EF_{EC} and EF_{OC} of open fire burning for wheat were 0.16 - 0.17 and 0.29 - 2.81 g/kg, respectively^{23, 31}. The difference is likely due to the restricted air supply and poor mixing in residential stoves compared with those in chambers and open field, resulting in relatively lower combustion efficiency and higher combustion temperature, which is favorable for EC formation⁵.

For coal combustion, EF_{EC} varied from 0.006 for the anthracite coal (Beijing, honeycomb) to 0.83 ± 0.34 g/kg for the MVB chunk coals from Taiyuan. The mean and standard deviation of EF_{EC} for coal was 0.23 ± 0.36 g/kg. Similarly, EF_{OC} ranged from 0.007 to 1.00 g/kg for these coals. These results were comparable to those previously reported. For example, Chen *et al.* found EF_{EC} measured in residential stoves ranged from 0.004 (anthracite) to 0.25 g/kg (MVB) for honeycomb briquette, and from 0.007 (anthracite) to 13.3 g/kg (MVB) for raw chunk coals⁷.

Size Distribution of PM₁₀ from crop residue and coal combustion

For all crop residues tested in whole burning cycle, the distributions were similar and unimodal with the peak between 0.7 and 2.1 μ m (S6). The similarity leads to small standard deviation of the overall distribution of all crop residues. On average, over 81% of the total mass of PM₁₀ from crop residues was PM_{2.5} and approximately 12% were finer particles with D_a less than 0.4 μ m (PM_{0.4}). Unlike crop residues, size distributions of PM₁₀ from 5 coals fell into two distinguished categories (S6). For chunk coal from Taiyuan and chunk coal A from Yulin, size fraction between 0.7 and 2.1 μ m contributed 49 ± 11% of the total mass of PM₁₀, while the dominant fraction of two honeycomb coals and chunk coal B from Yulin was those with D_a less than 0.7 μ m, accounting for 52 ± 18 to 60 ± 1% of the total. For all coals tested, PM_{2.5} fractions were more than 77 ± 5% of the total. The domination of fine particles from coal combustion emissions was often reported⁷.

Discussion

Difference in EF_{PM} among Crop Residues

Like those reported in the literature, the measured EF_{PM} varied widely among crops and coals. A number of factors including fuel property, stove type, oxygen supply, combustion temperature, and fire management have been investigated for their influences on EF_{PM} ^{13-23, 32-35}. For example, EF_{PM} for wheat and corn residues burned in household stoves ranged from 0.12 to 29.0 g/kg¹³. Dhammapala *et al.* found that PM_{2.5} emission from wheat stubble burning decreased from 4.7 ± 0.4 to 0.8 ± 0.4 g/kg when the combustion efficiency increased from 92.2 to 97.7%¹⁹. To address the factors affecting EF_{PM} for crop residues, a number of factors including the measured contents of moisture, C, H, and N of the fuels as well as the calculated MCE, PIC, R_b , and R_c were assessed using a stepwise regression model. The detailed result is presented in S7. Of these parameters, moisture and MCE were significant (p < 0.05) in smoldering phase, flaming phase, and whole burning cycle and 63 - 83% of the total variations in EF_{PM} can be explained by them. As such, the regression models can be applied to predict EF_{PM} based on moisture and MCE and the predictions are plotted against the measurements for the three experiments in Figure 1.

In smoldering phase, flaming phase, or whole burning cycle, moisture appeared to be the most important factor affecting PM emission. However, the influence of moisture on EF_{PM} was complicated as documented in the literature. For example, it was found that PM concentrations were 34.2, 161, and 70.8 mg/m³ from combustion of firewood with moisture contents of < 25, 26 - 39, and > 40%, respectively³⁵. The presence of water resulted in lower combustion efficiency, leading to a thick cloud of smoke particles³³. Slower formation and hence lower PM emission rate can also occur due to lower combustion temperature under higher moisture content¹⁷. In addition, steam stripping or volatilization of organic

compounds, which is fuel moisture dependent, can also result in the change of PM mass³⁶. In this study, significantly negative correlation (p = 0.004) between moisture and EF_{PM} was found. Since the crop residues were stored at the same condition for months prior to the experiment and the difference in moisture content was likely due to differences in fuel composition and texture. It will also be interesting to quantitatively test the influence of moisture on emission for the same crop residue in the future. The second important factor was MCE, which reflects the status of oxygen supply and combustion efficiency. It was found that similarly defined combustion efficiency explained more than 60% of variation in PM_{2.5} emission for wheat straw¹⁹. It was indicated that although MCE is affected by moisture and other fuel properties, it is also related to non-fuel factors, such as air supply and mixing during combustion²⁰.

Difference in EF_{PM} among Coals

Larger difference in EF_{PM} was observed between the honeycomb and chunk coals and even among the three chunk coals tested. It was reported that PM_{2.5} emissions from chunk coal combustion were 1.4 to 4 times of those from combustion of honeycomb made from the same coal²⁹. A number of parameters including moisture, ash content, VM, heat value, and MCE were tested for their influences on EF_{PM} for coals using a stepwise regression. It was found that the two most significant factors affecting EF_{PM} for coals were MCE (p = 7.0×10^{-7}) and VM (p = 0.0003) and 92% of the variation was accountable (S7). Figure 2 presents the relationship between the model-predicted and measured EF_{PM}. Higher EF_{PM} were also reported for coals with higher VM previously⁷. It is also known that bituminous coal generally ranks first in both VM content and EF_{PM} among various coals, followed by sub-bituminous and anthracite^{7, 29}.

Difference in Particle Size Distributions

As discussed previously, size distributions of PM₁₀ from burning of various crop residues were similar to one another (Figure S2). Still, it was found that the minor difference among crops was moisture dependent. Of the 9 size stages, correlation coefficients between moisture and relative fractions of 6 stages with D_a larger than 1.1 μ m were positive (5 out of the 6 were significant at p < 0.05), while correlation coefficients between moisture and relative fractions of the remained 3 stages with D_a less than 1.1 µm were negative (p =0.098, 0.041, and 0.054 for 0.7 - 1.1, 0.4 -0.7, and $< 0.4 \mu m$, respectively). Such a relationship was aggregately characterized by the significantly negative correlation between moisture and fine/coarse ratio (mass of PM smaller than 1.1 µm divided by that larger than 1.1 μ m) (r = -0.651, p = 0.002). A similar linear correlation between fuel moisture and PM mean diameter was also found in residential wood combustion³⁷. It is believed that the increase of moisture can reduce combustion temperature and efficiency³³⁻³⁵. Since larger particles are produced under lower combustion temperature¹⁷, higher moisture is favorable for emission of larger particles. Higher temperature may also shift mass distribution of particles to smaller diameter by limiting partitioning of organics on particles³⁷. In addition, fuel moisture may also affect relative humidity of flue gas, subsequently particle condensation in flue gas, and the size of new emitted particles²⁴. As EF_{PM} was also negatively proportional to moisture as discussed above, a negative correlation between EFPM and PM size was expected. The same relationship was also revealed in sawdust combustion¹⁷.

The size distributions of PM_{10} from five coals can be divided into two categories with dominant size ranges of 0.7 - 2.1 µm or < 0.7 µm (Figure S2). Of all coal properties and combustion status determined in the study, the only one which distinguished the two categories was Char Residue Characteristics (CRC, an index describing caking property of combusted coal residue, the higher the CRC the tighter the combusted residue)³⁸. The CRCs

of the 2 coals emitted PM_{10} with dominant D_a range of 0.7 - 2.1 µm were 5 and 6, while CRCs of the 3 coals emitted PM_{10} with dominant D_a range of < 0.7 µm was 1 or 2. It appeared that the coals with higher CRC had stronger caking potential and tended to emit larger particles during the combustion, primarily due to decrease in particle surface area and increase in contact time between the volatiles and char³⁸.

Difference in EF_{EC} among Different Crop Residues and Coals

 EF_{EC} from crop residues varied widely from 0.493 (peanut) to 2.63 g/kg (wheat). A larger difference in EF_{EC} among different crop residues with a coefficient of variation of 74% was also reported before²⁵. Besides fuel type, many other factors including combustion efficiency, air flux, burning rates, and fuel loading can also affect EF_{EC} ^{5, 25, 39}. In this study, five factors of MCE, moisture, N, C, and H were evaluated using a stepwise regression model for their influences on EF_{EC} . The details of the regression analysis can be found in S7. It was found that moisture and N were significant for EF_{EC} (p < 0.05) and can explain 63 - 72% of the total variations in EF_{EC} for whole burning cycle and two separated burning phases. A good agreement between the measured and predicted EF_{EC} using a regression model with moisture and N as independent variables is shown in Figure 3. It appears that low moisture were favorable for EC emission, as high moisture content can suppress combustion temperature²⁰. More studies are required for understanding the effect of N on EC emission, which can not be explained at this stage.

In coal combustion, the four significant factors (p < 0.05) identified include ash content, VM, heat value, and MCE, which contributed 95% of the total variation of EF_{EC} (S7). Of these factors, heat value was the most important one which can explain 48% of the total variation. The mechanisms of MCE and VM influence are similar to those for PM emission. Effects of VM and heat value on EC emission from coal combustion were also reported by other researchers^{5, 7} and it was suggested that VM in coal was associated with coal tar and hydrocarbons that serve as nuclei for EC formation⁵.

Relationship between EF_{EC} and EF_{PM}

Significant correlations among EF_{EC} , EF_{OC} , and EF_{PM} were found in this study (p < 0.05) as well as in the literature^{7, 15}. For various crop residues tested, EF_{EC}/EF_{PM} were 0.18 ± 0.06 , 0.19 ± 0.11 , and 0.19 ± 0.09 for flaming, smoldering and whole cycle burning, respectively, showing relatively small variation among crops and no significant difference among the stages (p > 0.05). This is also true for EF_{OC}/EF_{PM} (0.17 ± 0.06 , 0.19 ± 0.07 , and 0.19 ± 0.07 for the three experiments, respectively). The results generally agree with those reported by the others^{21-23, 30}. For the coals, EF_{OC}/EF_{PM} (0.18 ± 0.12) were similar to those of crop residues, while EF_{EC}/EF_{PM} (0.07 ± 0.06) were significantly lower than those of crop residues. Again, similar results have been reported previously^{7, 29}. The similarity in influencing factors on EF_{EC} and EF_{PM} also suggests the correlation between them.

EC is usually associated with fine and ultrafine particles in air. For crop residues studied, six fractions with size larger than 1.1 μ m were negatively correlated with EF_{EC} (p < 0.05), while the coefficients between EF_{EC} and the other three fractions were positive (p < 0.05). A significant linear relationship was revealed between log-transformed EF_{EC} and EF_{PM0.4} and the details can be found in the S8.

Future Study

All above discussion was based on the data collected in this study under given circumstance and can not be simply extrapolated. For example, open-field burning may have very different results and stoves used in rural China can differ from those used in other developing countries.

Taking the large variations of EF_{EC} and EF_{PM} into consideration, more studies are preferred for a better characterization of the emissions and a better prediction of emission inventory. For laboratory studies, influences of various factors including type and condition of stoves, type and property of fuels, burning and environmental conditions, and the way of burning should be investigated systematically. The results should be compared and validated using filed observed data of relatively large sample size. Based on these results, emission models can be further improved. Globally, more studies on stove emission in developing countries other than China are also important.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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Figure 1.

Comparison between the measured and calculated EF_{PM} for crop residue burning. The calculation was based on regression models with two independent variables of moisture and MCE. Three experiments including smoldering phase, flaming phase, and whole burning cycle are presented together.



Figure 2.





Figure 3.

Comparison between the measured and calculated EF_{EC} for crop residue burning. The prediction was based on two independent variables of moisture and N. Three sets of experiments presented include smoldering phase, flaming phase, and whole burning cycle.