Reply to the comments of anonymous reviewer #2 on manuscript entitled " Emission characteristic of refractory black carbon aerosols in the fresh Asian biomass burning: a perspective from laboratory experiment "

We appreciate very much the insight comments and recommendations of the reviewer in improving this paper and our future research. Here, we will response to all the comments one by one as follows:

This manuscript reports results of rBC emissions and emission ratios from two different agricultural fuel types, which can be emitted during open agricultural burning and are relevant to both China and other regions where wheat and rapeseed are grown worldwide. The authors do a good job of citing previous work on ambient rBC emissions and the large uncertainties and variation in the data from different locations and sources. Unfortunately this work only focuses on rBC and does not measure non-rBC mass or aerosol optical properties. A large source of uncertainty in the aerosol optical properties from OBB is due to the presence of BrC as well as rBC, which the authors acknowledge in the introduction, but do not attempt to measure or quantify. For example, even with the assumption that the non-rBC aerosol is dominated by organics (and not direct measurement), this work could predict total aerosol optical properties and BrC absorption using the Saleh et al. 2015 (already cited) and Pokhrel et al. 2017 correlations of mass ratios with measured aerosol optical properties from different fuel types during a similar laboratory study on different biomass burning fuel emissions. A measurement of the total aerosol, Organic Aerosol and/or non-refractory aerosol mass to report rBC/OA or rBC/Total aerosol mass is used to bound BrC as referenced above as well as the total aerosol optical properties (single scatter albedo, SSA) that has been found to also be independent of fuel type and as a function of MCE. SSA is relevant to the total aerosol radiative impacts of OBB as reported Liu et al., 2014. For all of these reasons, the addition of non-rBC measurements if available for this data set would greatly enhance the impact of this work on the total aerosol optical and physical properties from near-field source emissions of two major crops from China, wheat and rapeseed, and any attempt to add this kind of total aerosol information if possible would be greatly supported.

Reply: We consented to the reviewer's comments that open biomass burning (OBB) emits not only refractory BC (rBC) particles but also substantial mounts of organic aerosols (OA), the latter of which mostly consists of light-absorbing carbon (BrC). The chemical/physical and mixing state of BrC with rBC were not well understood, which resulted in great uncertainty in evaluating the climate effect of biomass burning aerosol. Recent studies (Saleh et al., 2014, Pokhrel et al., 2017) make advances in better quantify the absorptivity and direct radiative effect of BrC mass by introducing an applicable parameter (BC-to-OA ratio)(Saleh et al., 2015). They also revealed that absorptivity of BrC depend largely on its combustion condition at the source (Saleh et al., 2014, Collier et al., 2016), reflecting of the importance to investigate the optical properties of fresh biomass burning aerosols. In the present study, initial idea is to investigate the emission characteristics of rBC from open burning of agriculture residues in East China to better constrain uncertain of rBC emission inventory and to improve the performance of regional chemistry model, because intensive crop residues burning in the field often resulted in local Air Quality Index > 500 (hazardous level) in harvest season in East China. Because of the limit in instruments, the optical property of OBB aerosol was not simultaneously measured during experiments. Nevertheless, we would like to follow the reviewer's advice to incorporate optical measurements of OBB-related aerosols in our further research.

Regarding reviewer's suggestion to predict BrC absorption on the basis of assumption that the non-rBC aerosol is dominated by organics and BC-to-OA ratio. Note that, SP2 only counts the number concentration of light scattering particle or non-rBC particles with diameter larger than 166 nm due to limitation of avalanche photodiode detector, and the size of non-rBC particles are derived on the basis of predetermined calibration curve using polystyrene sphere latex particles (PSL, Size Standard Particles, JSR Corporation, Japan). Therefore, precise estimation on optical properties of organics aerosols was difficulty due to lacking the size-resolved density, morphology information. In particular, the information about existing state of non-rBC mass (internally mixed or external mixed with rBC) for the particle with a diameter less than 166 nm was also unknown. As a whole, it is better to predict the optical property of OBB aerosol on the basis of direct measurement. Now we are purchasing a centrifugal particle mass analyzer (CPMA), we will use a DMA-CPMA-SP2 tandem system to quantify the mass concentration of rBC and non-rBC, as well as the optical properties of rBC-containing particles. By selecting a specific particle size range (for example, 200 ~ 300 nm), the mixing state and absorptivity of BrC will be quantified properly.

General comments:

The rBC sample was diluted by a factor of 46 while the gas-phase measurements were diluted by a factor of 22. There a concern that such a large dilution of the rBC would make quantification of the total rBC mass from the fire emissions have very large uncertainties in the measurements. A study of the uncertainties induced by the dilution system was studied for the aerosol sampling, but was not quantified for the gas-phase measurements. Are the authors not concerned that the different dilution ratios for the aerosol measurements and the gas-phase measurements might not introduce uncertainties in the measurements as the emissions ratios are the main deliverable of this manuscript?

Reply: We agree with reviewer that different dilution factor could result in different uncertainties in both aerosol-phase and gas-phase measurement, and finally influences the emission ratio of rBC. During biomass burning experiment, we used a high dilution factor (~46) for aerosol channel to satisfy the maximum detection limits of SP2. As a matter of fact, the same dilution factor (46) was also examined for gas-phase measurement. We found that uncertainty (2%) of carbon monoxide measurement was negligible. Because an ultrafast CO analyzer (model AL5002, Aero- 30 Laser GmbH) has relatively high detection limit (detection range 0–100 ppmv, detection limit 1.5 ppbv, integration time 1 s), we selected a low dilution factor (22). Besides, the inlet of gas-phase and aerosol-phase sampling tube was ~40 cm beside the flame of biomass, therefore except for 46 times excess dilution, biomass smoke has already underwent great dilution at the sampling location due to mixing with excess fresh air. In this study, we considered that the emission ratio of rBC (ΔrBC/ΔCO) preserved because they were emitted in seconds and they did not experience in-cloud and below-cloud scavenging processes. In future work, we will follow reviewer's suggestion to use the same dilution ratio when both rBC and CO concentrations were measured.

It is also unclear as to why the authors did not conduct dilution studies to see if the rBC coating was changing by introducing a 1/46 dilution ratio. Sampling a range of initial fuel sample emissions including smaller burn sizes (< 20 g) from the same fuel types would have greatly enhanced this study.

Reply: We agree to the reviewer's suggestions. As implied, different dilution condition will impact the gas-aerosol equilibrium of semi-volatile organic matter, the latter of which tend to influence the mixing state of rBC particles due to continuous condensation/evporation processes. In this study, we did not quantify the effect of dilution on the coating thickness of rBC particles because the total dilution factor can not be exactly determined since the sampling inlet was besides the combustion flame, and biomass smoke has already diluted due to mixing with fresh air in the combustion chamber. Besides, to avoid saturation of SP2 measurement, the burning biomass was only ~20 gram, and the rapid evolution of the combustion process also resulted in difficulty in discriminating the dilution effect. Therefore, large-scale burning experiment (like FLAME experiments) is much applicable for investigating dilution effects, and parallel measurements using two SP2 with different dilution factors is another option. In our future research, we would like to follow the reviewer's advice to investigate the dilution effect.

What is the width of the rBC size distributions from each experiment? While MMD of rBC MED is reported for each experiment, for example, what is the sigma or range in rBC size distributions? Is the rBC a tight size distribution/at what diameter does the rBC drop off for both the high and low ends? A table with this information and/or a graph of the rBC size distribution averages or examples would make a good addition to this manuscript in understanding the size range of rBC emissions.

Reply: Great thanks for the reviewer's suggestions. We will add the information in the table 2. Besides, We make a correction in the curve fitting of mass size distribution of rBC particles according to the comments of the reviewer #1. As a matter of fact, Lognormal function fitting is better than that of Gaussian function in most instances. This phenomena is more prominent in smoldering-dominant case that that in flaming-dominant cases (as shown in Figure 1). We will correct the expressions in the revised manuscript, and add dM/dlogDp plot in the Figure 2a.

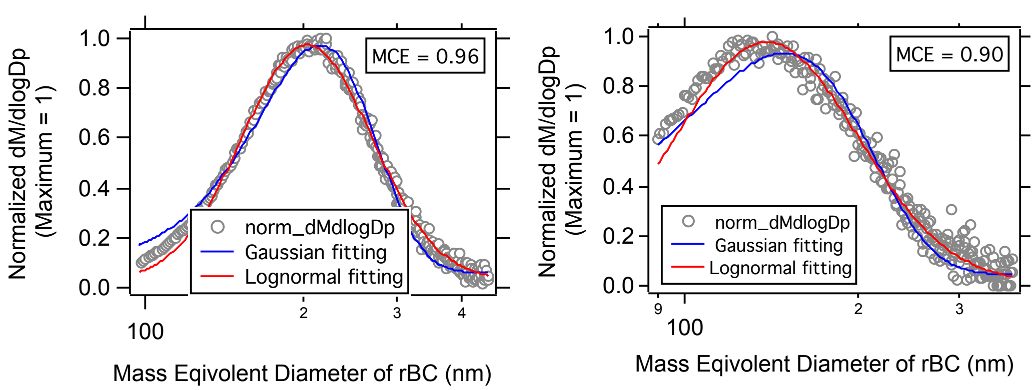

Figure 1 Gaussian and lognormal fitting for normalized mass size distribution of rBC for flaming- (left) and smoldering- dominant case (right).

Could the data from each burn be separated into smoldering and flaming analysis? What was the reason for using a fire-integrated MCE analysis when the first Figure separates the different phases? How was the separation of combustion conditions done for that figure? Was it using 0.9 as referenced in the introduction (Page 3 Line 10-11) or 0.95 in the results section (Page 11 Line 2) to separate the phases (or some- thing else)? This information is referenced in different ways in the results section, but should be clearly defined in the experimental section and remain constant throughout the results section (which it may be but it's not clear to the reader how this was done).

Reply: As mentioned in the manuscript, the combustion processes evolved rapidly. Flaming and smoldering combustion sometimes occurred at the same time at different part of biomass, and clear discrimination between flaming phase and smoldering phase was difficult. Therefore, we used a fire-integrated MCE to represent the general burning condition of biomass. Regarding the criterion, a value of MCE > 0.95 and a value of MCE < 0.9 was used in literatures as criterion to separate flaming and smoldering phase, respectively. We followed the same criterion in the manuscript. Figure 1 shows two examples reflecting the temporal variations of number concentrations of rBC and non-rBC particles. We found a prominent phenomenon that occurrence of peak of number concentration of non-rBC particles was obviously later than that of rBC particles, and the MCE value = 0.95 always fall in the middle of these two peak. It can be an indicator that the combustion shifted from the flaming-dominant to smoldering-dominant state. We will clearly state the MCE criteria in the revised manuscript.

In the absence of other size distribution or measurements of the non-refractory or scattering aerosol, if this SP2 is able to measure scattering particles up to 1 micron in diameter, could the scattering data be presented in addition to the rBC data to give a more representative picture of the total aerosol emissions and optical properties?

Reply: As suggested by the reviewer, we would like to add size-distribution information about both the rBC and non-rBC particles during the burning experiment, as shown Figure 2 below. SP2 only detects light-scattering or non-rBC particles with diameter larger than ~ 166 nm. Presuming that non-rBC particles were in spherical configuration, the volume size distribution could be also determined. In the present study, non-rBC particles less than 166 nm in size (the lower detection limit of the SP2) accounted for limited fraction (4%) of the total aerosol volume. The maximum size of non-rBC particle was estimated to be ~ 750 nm. It is worth noting that we found that there were two modes for the volume size distribution of non-rBC particles with a peak at 307 nm and another peak at 606 nm. The second minor peak was related to an extreme smoldering case (discussed in paper, Inomata et al., 2015, JGR), since the number concentration was very low.

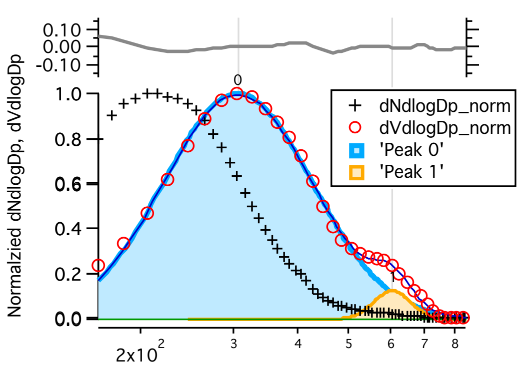

Figure 2 Normalized number size distribution (dN/dlogDp) and normalized volume size distribution (dV/dlogDp) for non-rBC particle during experiment.

Similar to the response of the first reviewer, the authors focus on the combustion state influencing the rBC emissions. What about the effect or concern for differences due to different fuel types, e.g. agricultural fuels versus wildfire fuels? Fuel types vary largely for OBB, and this should not be neglected. The authors are advised to modify the interpretation of the results *and text at times to accommodate this as another reason for the large variability in reported rBC emissions from both laboratory and ambient measurements.*

Reply: We consented to the reviewer's insight comments. To avoid misleading, we will specify the type of biomass in the table 3 and correct our statements in the revised manuscript. As the reviewer implied, there are large differences in carbon/water contents and physical structure for different biomasses, which may result in their different inflammability, as well as mixing state of rBC particles, though some studies (e.g. Collier et al., EST, 2016) pointed out that general characteristics of biomass burning aerosols depended strongly on the combustion processes of a fire, and Saleh et al., (2014, NG) reported that aerosol absorptivity depends largely on burning condition, not fuel type.

The addition of the wet data needs further substantiation in the methods, focus in the text, and data interpretation. Without this it should be removed from the text (or alternatively moved to SI).

Reply: As suggested, we will move this part to supporting information.

Specific Comments:

Page 1, Line 14: Are "rape plants" the same as rapeseed? If so, suggest adding"also known as rapeseed" to the text.

Reply: We will use rapeseed plants in the context for consistency.

Page 1, Line 15: Do the authors mean "used" when they say "adopted"? Adopted implies a change was made to the standard SP2 rBC sampling regime. If this was done, please state and explain, and if it was not, please change the text to "used" or similar wording as the SP2 is a standard instrument for measuring rBC.

Reply: In the revised manuscript, we will replace "adopted" by "used".

Page 1, Line 1 – Page 2, Line 1: "This study highlights that open biomass burning produces the majority of coated rBC particles, which have considerable ability to affect cloud processes and influence regional climate." This significance statement in the abstract overstates the results reported in this paper. It is unclear how the authors can state that biomass burning produces the majority of rBC coated particles in the atmosphere from a laboratory study of two different agricultural fuel types. A similar statement could be made along the lines of agricultural fuel types produce coated rBC particles, . . .", which would not over interpret the reported results.

Reply: As suggested, we will revise the overstatement in manuscript, as follows: " This experimental study found that mixing state of rBC particles from biomass burning strongly depend on the its combustion processes, and overall MCE should be took carefully into consideration while the climate effect of rBC particles from open biomass burning was simulated."

Page 2, Line 3-4: What about cloud albedo?

Reply: We will add "cloud albedo" in the sentence.

Page 2, Line 7: It would be helpful to define OBB since this is not common terminology for a general audience. The reviewer suggests defining OBB to include agricultural and wildfire emissions, but mainly suggests adding a sentence to define OBB clearly to the reader.

Reply: we will add a terminology in the manuscript.

Page 2, Line 9: Do the authors mean VOCs or SVOC's? Both are common terms and are not used interchangeably.

Reply: Thanks for pointing out the typo, and I will delete the "VOCs" since it is not used in the context.

Page 2, Line 10: Suggest removing "in smoke"

Reply: "in smoke" is removed in the revised manuscript.

Page 2, Line 19 – 20: "Hygroscopic growth of rBC-containing particles also results in much more compact rBC cores." Is there a reference to support this statement? Suggest moving the Fan 2016 reference to a modelling study in the following sentence here and at the beginning of the sentence adding "Modelling studies indicate that the . . . " unless a measurements reference can also be added to support this statement.

Reply: We revised the sentence as suggested, as follows: " Modeling study indicated that hygroscopic growth of rBC-containing particles also results in more compact rBC cores (Fan et al., 2016). "

Page 2, Line 21-22: "Second, the rBC particles are often located at off-center positions or may possibly be attached to the surfaces of non-rBC particles." Are there any references that can substantiate this sentence? If none can be found, please remove this sentence or change it to a statement implying these morphologies are possible but not implying the significance of off-center rBC particles in ambient aerosols.

Reply: The sentence will be changed to "Second, the rBC particles sometimes were also reported to be attached on the surface of non-rBC matters (Moteki et al., 2014)."

Page 2, Line 28: add Liu 2015b to the list of citations for BrC influencing overall rBC absorption enhancements.

Reply: I will cite Liu et al., 2015b in the manuscript.

Page 3, Line 4-5: Suggest relating tar balls to secondary organic aerosol SOA from BB sources to link the two terminologies. Are tar balls one type of SOA defined as being low volatility and from BB sources? Are there any known optical properties that can be ascribed to this particle type, e.g. likely to contain BrC? A brief summary/explanation of the definition of what a tar ball is in terms of its formation, sources, physical and optical properties would benefit a larger audience.

Reply: In the revised manuscript, we will state that " It was reported that "tar-ball" that mostly consisted of BrC and secondary organic aerosols with low volatility were also emitted during smoldering combustion (Pósfai et al., 2004)."

Page 3, Line 12: When defining the rBC emission ratio, rBC is rBC mass concentration, correct? Likewise CO is a mixing ratio? Suggest adding this information to the initial definition here.

Reply: Initial definition will be added in the revised manuscript, as folows: "The rBC emission ratio (ΔrBC/ΔCO), which is defined as the enhancement of mass concentration rBC (in unit of ng/m3) with respect to its background versus that of CO (in unit of ppbv, parts per billion volume), ... "

Page 3, Line 27: "The variability in ΔrBC/ΔCO among observational studies also result from differences in sampling locations and conditions." After this section would be a good time to introduce the topic of variability due to fuel type as suggested in the general comments.

Reply：I will follow the reviewer's suggestion to revise the content of second paragraph in page3, as follows: "The rBC emission ratio (ΔrBC/ΔCO), which is defined as the enhancement of mass concentration rBC (in unit of ng/m^3) with respect to its background versus that of CO (in unit of ppbv, parts per billion volume), is an applicable indicator for constraining the rBC emission inventory for models (Pan et al., 2011). The variability in ΔrBC/ΔCO among observational studies mostly results from differences in measurement techniques, fuel type, and burning conditions. For example, observations made onboard the NOAA WP-3D aircraft yielded ΔrBC/ΔCO values of 9 ± 2 ng/m^3/ppbv (Spackman et al., 2008) and 17.4 ng/m^3/ppbv (Schwarz et al., 2008) for brush fire plumes during the TexAQS field campaign. Airborne observations on the NASA DC-8 aircraft indicated that the ΔrBC/ΔCO values were 8.5 ± 5.4 ng/m^3/ppbv for plume of boreal forest and agriculture fires in Asia, and 2.3 ± 2.2 ng/m^3/ppbv for wildfire plume in North America (Kondo et al., 2011). Observations using a multi-angle absorption photometer (MAAP, which employs the filter-based light absorption technique; here we consider BC instead of rBC) at mountain sites (30.16°N, 118.26°E, 1840 m a.s.l.) in South China yielded high ΔBC/ΔCO values (10 - 14 ng/m^3/ppbv) when the site was subjected to burning of crop residues (Pan et al., 2011)."

Page 4, Line 19 – 20: Suggest removing "to preserve its original state" since while this storage would limit deposition onto the samples it would not preclude semivolatile evaporation and/or water loss etc.

Reply：As suggested, the statement "to preserve its original state" will be deleted. As a matter of fact, the farmer normally dries agriculture residues in the sun for days before the biomasses are burned. The samples that we collected in the field were dehydrated so that evaporation of semi-volatile and water vapor should be negligible.

Figure 3 Photo of dry rapeseed plants that were collected in the field.

Page 4, Line 26: "To monitor the evolution of the combustion process of biomass, the mixing ratios of CO2 and CO in the OBB smoke were measured simultaneously." The gas phase mixing ratios were measured "to monitor the combustion conditions" of the rBC, correct? The statement here seems to imply aging, which these experiments are more representative of near-field emissions and do not probe aerosol aging in the plume as might be interpreted with the current sentence. The reviewer also cautions the authors to imply that all fires proceed from flaming to smoldering combustion conditions both here and other locations in the text since OBB can vary over time and does not always proceed as straightforward as a laboratory study.

Reply: We agreed to the reviewer's comments. Here, mixing ratios of CO and CO2 were measured to calculate Modified Combustion Efficiency (MCE), the latter of which is a applicable metric to estimate the combustion phase of biomass. To avoid misleading, we will revise the statement in the revised manuscript, as follows: "As mentioned, MCE is a useful metric for describing the combustion phase of biomass burning, and the calculation of MCE requires simultaneous measurements of CO and CO2 concentration. Here, mixing ratio of CO2 was measured using a Li-7000 CO2 analyzer..."

Page 5, Line 31-33: Suggest showing at least an example of the rBC size distribution from one or an average from several burns ideally as a figure in the main text, and alternatively in the SI. Is a Gaussian fit best or does lognormal fit the rBC mass equivalent diameter data better?

Reply: We will add the mass size distribution of rBC particle in the manuscript. We found that lognormal curve fitting is slightly better than that of Gaussian function for rBC particles in most burning instances, in particular for smoldering-dominant case, as shown in Figure 4. We will correct the statements.

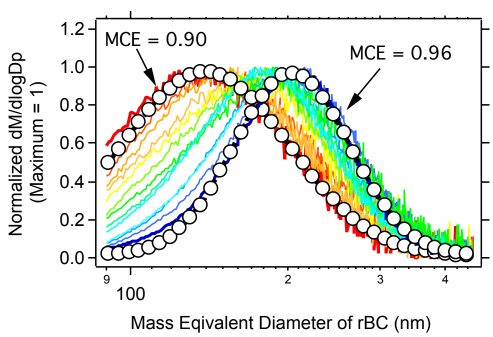

Figure 4 Normalized mass size distribution of rBC particles for all the burning cases.

Page 6, Line 3-4: The SP2 scattering channel was not saturated for the 500 nm and 1000 nm PSL's? The lower limit of the scattering detection is listed as 166 nm. What is the upper limit for this instrument? If this SP2 can detect scattering particles up to 1 micron in diameter, it would advantageous to report this data in addition to the rBC measurements.

Reply: Great thanks for the reviewer's suggestion. Regarding calibration, scattering signal for PSL particle at both Dp = 500 nm and Dp = 1000 nm were saturated for APD of SP2. Nevertheless, peak height of scattering signal for PSL particle at Dp = 500 nm could be estimated properly according Gaussian fitting by SP2. Unfortunately, it was not available for

PSL particle at Dp = 1000 nm due to strict data screening. Maximum size of non-rBC particle was estimated to be ~ 750 nm, and the We will report the information in the revised manuscript.

Page 8, Line 10: Fire-integrated MCE's are reported and listed in Table 2. What is the variability over the course of each burn? Could the range of MCE's from each experiment also be included in this table?

Reply: We will include 10th, and 90th percentile values for each experiment.

Page 9, Line 8-10: Could this be related to how the burns were started? Information on the fires were started/lit should be added to the information in the experimental section as well as being considered as an potential explanation for this initial rBC peak in number at the start of sampling.

Reply: We will add information about the ignition of biomass in the experimental section, as suggested.

Page 9, Line 22: ". . . because the combustion process differed significantly." Does this imply that the experiments do not generally proceed from flaming to smoldering as well as the examples in Figure 1? Please explain what this sentence means as it was not clear to the reviewer.

Reply: We will revise the expression to avoid misleading. In general, the combustion proceed from flaming to smoldering for most case, however the time duration of in flaming or smoldering were different. We will also add the total duration of combustion processes in the Table. Of course, we cannot avoid the situations that both flaming and smoldering combustion occurred at different position of fuel at the same time. Therefore, we considered using a fire-integrated MCE to present the overall combustion condition.

Page 9, Line 24: "45 times dilution". Earlier this was stated as 46 – please explain the reason for the difference.

Reply: We will correct the mistake.

Page 9, Line 27-29: Reference is made to the rBC displaying "a perfect Gaussian distribution for all burning cases." Reference is also made to a Figure 2a, which appears to not exist in this version of the text. This size distribution information should be added to the Figure. Is the rBC distribution averaged over the course of the whole experiment? rBC distributions are not often perfect Gaussians, therefore, the addition of this information to the Figure should be included.

Reply: As suggested, we will include the mass size distribution of rBC particles as shown previously.

Page 9, Line 31: Change " tends to produce larger particles" to "tends to produce larger rBC particles".

Reply: We will change to "tend to produce larger rBC particles"

Page 10, Line 2: "small" seems to contract the data in Figure 2 and previous sentence since the rBC MMD increases for flaming combustion.

Reply: Sorry for misleading, I will revise this statement as follows: " This result indicates that flaming combustion tends to produce larger rBC particles than smoldering combustion. It is consistent with previous studies, which reported that rBC particles formed considerably in intense flaming combustion due to less efficient transport of oxygen into the interior flame zone. As a result, growth in the size of rBC particles was rapid because the coagulation rate of particles is roughly proportional to the square of their number concentration (Lee and Chen, 1984)."

Page 11, Line 1 – 4: But are these reported differences in delta rBC/delta CO statistically significant? Based on the uncertainties, there does not look to be enough difference within the uncertainties of the measurements to warrant significant difference and subsequent interpretation.

Reply: We will add more information and revise the manuscript as follows: "the average ΔrBC/ΔCO ratio was 13.9 ± 10.1 ng/m^3/ppbv for the burning cases with a fire-integrated MCE > 0.95. This value was probably a low estimation since both flaming and smoldering combustions were included in the calculation. However, by selecting the cases with both the 10th and 90th percentiles MCE value > 0.90, we found that the average ΔrBC/ΔCO ratio was 23.1 ± 11.4 ng/m^3/ppbv ...".

Page 11, Line 29-30: What is the reason for focusing on the time delay analysis data when the LEO-fitting coating thickness analysis that yields more information with fewer uncertainties was also extensively done? Was the LEO-fitting analysis only done on the MED = 200 rBC core-containing particles? 200 nm MED is relatively large for most rBC studies, and even for some of the data presented here where MMD's are reported down to 144 nm MED rBC for some of the experiments. How much do the results change if 150, 180 or 220 MED cores were used for the LEO analysis?

Reply: As pointed out by the reviewer, LEO-fitting analysis could provide direct information about the coating thickness of rBC particle. Nevertheless, the delaytime of occurrence of incandescent peak *after* scattering peak also was informative for the reader to understand the real situation when the particle was passing through the laser. For instance, number of previous studies (Sedlacek et al., 2012, GRL; Moteki et al., 2007, GRL; Taketani et al., 2014, JGR, Miyakawa et al., 2015, AE, etc.) directly used the delaytime to indicate the degree of aging/coating of rBC-containing particles. rBC-containing particle in non-shell-core structure could also be determined if the delaytime value was a negative value (Sedlacek et al., 2012).

Regarding the LEO-fitting analysis, we agree with the reviewer's concerns that most of rBC particles had small size (less than 200 nm), and coating thickness could be calculated on the basis of LEO-fitting method. It was worthy noting that low detection limit of scattering particles was ~ 166 nm, implied that information about the rBC-containing particles with a diameter less than 166 nm was missing, and it will be problematic statistically to identify the relationship between coating thickness and MCE. Although we did analysis for all the data, we just reported the rBC particles with larger MED in the manuscript.

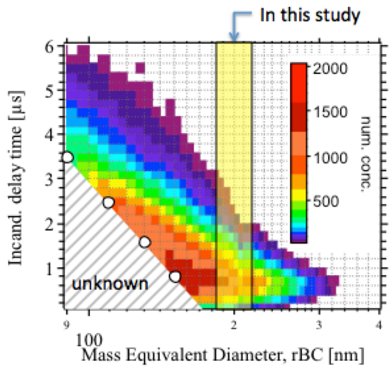

Figure 5 An example of variation of delaytime as a function of MED of rBC. Line-shaded area indicates that the information is missing. The yellow-color shaded area indicate the data that was analyzed in the manuscript.

Page 12 and Figure 4: Can you separate the data into flaming and smoldering to substantiate the claim that the two modes present in all the data are due to the different combustion phases?

Reply: In this study, we found that S/C ratio for rBC particles with MED = 200 nm increase as MCE decrease in most of burning case (Figure 6 shows two examples). As demonstrated, S/C ratio was ~ 1.2 when MCE > 0.95 (flaming-dominant phase) and ~ 1.4 when MCE < 0.90 (smoldering-dominant phase), in accordance with the estimated results from multi-Gaussian curve fitting. Because the data in flaming and smoldering phase overlapped significantly (as Figure 4 shown in the manuscript), it is better to demonstrate the differences using multi-Gaussian curve fitting method. We will add more information in the Figure 4 in the manuscript for better understanding of readers.

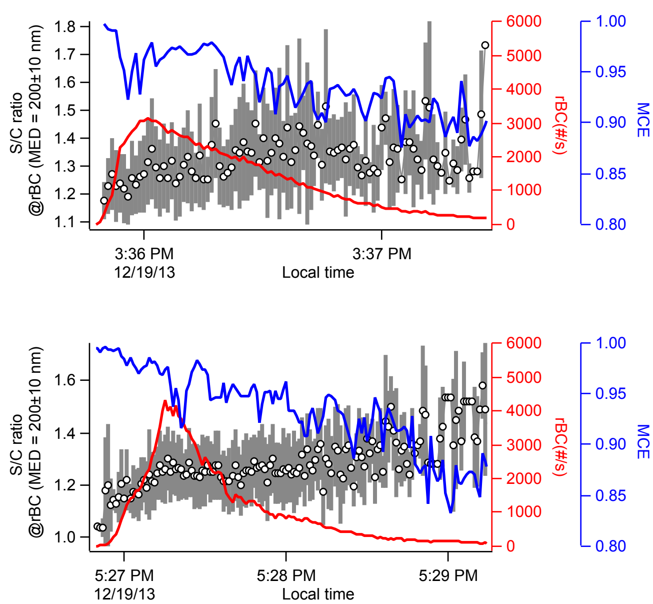

Figure 6 Two examples demonstrating that S/C ratio increase as MCE decrease for rBC particles with MED = 200 nm during combustion process.

Page 13, Line 6-13: Add the range of S/C reported for the ARCTAS data to the text. How much weight can be placed on a S/C change of 1.2 to 1.4? Could some of the other studies help to substantiate why this is a significant difference? More explanation and reference to other datasets here in the text would be advised since the data in Figure 5 appears to have a lot of scatter in the data and poor r2 fit values.

Reply: We will report the range of S/C ratio from ARCTAS data, and revise the statement as follows " Airborne measurements during the ARCTAS campaign (Kondo et al., 2011) showed an increasing tendency for the values of the S/C ratio (1.3 ~ 1.66) with increase of MCE, and the authors explained that this phenomena was because flaming phase plumes were more aged than the smoldering plumes, a 205 ± 40% increase of the volume of the coating materials resulted in a larger S/C ratio than that of rBC particles in smoldering plume."

We consented to the reviewer's comments that a S/C ratio increasing from 1.2 to 1.4 was not significant. As a matter of fact, the volume of non-rBC coatings could increase by 140% when S/C ratio increases by 20%, which will possibly lead to mass ratio of rBC to non-rBC (M_R = $m_{non\text{-}BC}$ /m_{rBC}) around 2.7. Recent study (Liu et al., NG, 2017) reported that the absorption enhancement due to optical lensing effect was significantly important at M_R = 3 for biomass burning aerosols. Liu et al., (2014, GRL) also reported that MCE could explain 60% of variability in single scattering albedo of biomass burning aerosols. It implies that the moderate variation in mixing state of biomass burning aerosol may have significant effect on its optical properties. As suggested by the reviewer, we will perform the optical measurement of rBC particle from biomass burning in our next research.

Liu, D., et al., (2017), Black-carbon absorption enhancement in the atmosphere determined by particle mixing state, Nature Geosci, advance online publication, 10.1038/ngeo2901

Liu, S., et al. (2014), Aerosol single scattering albedo dependence on biomass combustion efficiency: Laboratory and field studies, Geophys. Res. Lett., 41, 742–748, doi:10.1002/2013GL058392.

Page 13, Line 14 – 15: Is this for all the data? Is there a difference in the different fuel types or MCE flaming versus smoldering conditions if the data were to be averaged from all experiments and separated into different categories? Adding the range of thicknesses sampled should also be added to the text here as the coating thicknesses look to cover the full ranges reported by the aircraft data referenced in the text.

Reply: Yes, histogram analysis of coating thickness was performed for all the data. We found that it shows a lognormal distribution with a mode value of 20 nm and the standard deviation of 0.54. Coating thickness was mainly ranging from ~ 11 nm to 54 nm. The same analysis was also done for rBC particles with MED = 250 nm, what the difference was that histogram showed a Gaussian distribution with a mode value of ~17 nm. We will add the information in the revised manuscript.

Page 13, Line 19 – 29: Since Figure S4 indicated coating thicknesses of 0 – at least 60 nm sampled from this data, is it possible to state that atmospheric aging results in increased rBC coatings? The data presented here and in Table 3 is from a large variety of fuel types, combustion conditions, and atmospheric aging timescales that this level of interpretation requires more investigation isolating different fuel types and atmospheric aging timescales.

Reply: We consent to the reviewer's comment that it was hardly to attribute the difference in coating thickness of rBC particle among studies only to atmospheric aging because fuel types, combustion condition were different. We will add type of biomass in the Table 3 and revise the interpretation in the revise manuscript. Besides, we noticed that, for the rBC particles outflowed from urban area, the number fraction of thickly-coated rBC particles (S/C ratio > 2 @ rBC_{MED} = 180 nm) could increased from 30% to 60% of total rBC particles as their photochemical age increased from 2h to 14 h (Moteki et al., 2007). It reflected that the photochemical process was of great importance in variation of the mixing state of rBC particles. At present, more field/airborne observational evidences of biomass burning aerosols were needed to explicit the variability in literature.

Page 14, Line 4-5: The S/C ratio appears to increase with the EF of the NMVOC's for dry data while the wet wheat S/C does not look to depend on EF of NMVOC's. It also looks as if the S/C for the wet data is at the maximum for the dry data. More discussion of these differences should be included in the text if this is found to be significant. If not, then the wet data should be removed from the manuscript as it does not have much interpretation of the data collected here anywhere else in the manuscript.

Reply: We will delete the data from burning of wet wheat, as suggested by the reviewer.

Page 14, Line 9-10: Without a reference for this hypothesis or substantiation from the data presented here, this should be removed as it is too speculative.

Reply: We will remove the sentence from the manuscript.

Technical Corrections:

Page 2, Line 3: remove "the" from ". . . play a vital role in the climate change. . ."

Reply: "the" is removed in the sentence.

Page 2, Line 5: remove "their" from "its their"

Reply: "their" is removed in the sentence.

Page 2, Line 11: remove ", evidently"

Reply: "evidently" is removed in the sentence.

Page 2, Line 19: remove "much"

Reply: "much" is removed in the sentence.

Page 3, Line 27: "The variability in $\Delta rBC/\Delta CO$ among observational studies also result from differences in sampling locations and conditions." – change "result" to "results"

Reply: "result" is replaced by "results".

Page 4, Line 10: Change ". . . we conducted open burning experiments. . . " to ". . . we conducted laboratory burning experiments. . . ".

Reply: To avoid misleading, we replaced "open" by "laboratory" here and all other parts in the manuscript.

Page 4, Line 20: Remove "generally"

Reply: "generally" is removed in the sentence

Page 5, Line 32: change "fitted" to "fit"

Reply: "fitted" is replaced by "fit".

Page 13,Line 9: Remove "As a matter of fact"

Reply: "As a matter of fact" is removed in the sentence.

Page 14, Line 5: Change "open" to "laboratory"

Reply: "open" is changed to "laboratory" in the manuscript.

Page 14, Line 13: Remove "urgently"

Reply: "urgently" is removed in the sentence.

Page 14, Line 26: Remove "obviously"

Reply: "obviously" is removed in the sentence.

Page 14, Line 27: Add "rBC" to say "result indicated that the rBC coagulation/growth. . ."

Reply: "rBC" is added here.

Table 1: Change "C/S ratio" to "S/C ratio"

Reply: "C/S ratio" is corrected to "S/C ratio"

Table 2: Should the last column say "MED of rBC MMD"?

Reply: MED is abbreviation of Mass Equivalent Diameter, and MMD is Mass Mode Diameter, we will add annotation in the Table 2.

Figure 1: Needs the information added to the figure or figure text on the different color blocks of data shown in yellow, red and blue.

Reply: The information about the color blocks in yellow, red and blue is add in caption of Figure 1.

Figure 2: Needs an explanation of the lines presented in the figure and the circle around one set of data. Also the text refers to 2a and 2b within the Figure which are not present.

Reply: We will add the explanation of the lines and circle in the figure. The missing figures 2a and 2b will be added in the manuscript.

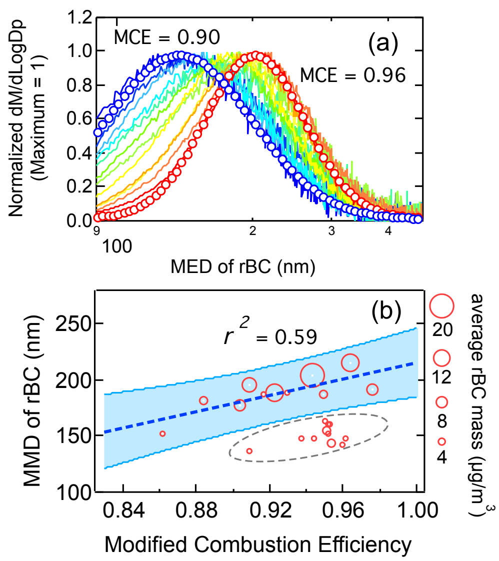

Figure 4: Would changing the color scale on the number of rBC in the Figure enhance the ability for the reader to discern the two modes that can be separated with the histograms? Label the modes flaming and smoldering on the figure would also make the main points of the Figure more clear to the reader.

Reply: We will polish the Figure 4 as suggested by the reviewers.