

Interactive comment on “Emission characteristics of refractory black carbon aerosols from fresh biomass burning: a perspective from laboratory experiments” by Xiaole Pan et al.

Anonymous Referee #2

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Summary:

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 ex-

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ample, 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.

References: Saleh et al. Contribution of brown carbon and lensing to the direct radiative effect of carbonaceous aerosols from biomass and biofuel burning emissions. JGR-Atmospheres, 2015.

Pokhrel et al. Relative importance of black carbon, brown carbon, and absorption enhancement from clear coatings in biomass burning emissions. ACP, 2017.

Liu, S. et al. Aerosol single scattering albedo dependence on biomass combustion efficiency: Laboratory and field studies. GRL, 2014.

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 is 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

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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? 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.

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.

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 something 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).

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?

Similar to the response of the first reviewer, the authors focus on the combustion state

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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.

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).

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

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.

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.

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

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

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wildfire emissions, but mainly suggests adding a sentence to define OBB clearly to the reader.

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

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

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.

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.

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

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.

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.

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Page 3, Line 27: "The variability in $\Delta rBC/\Delta CO$ 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.

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.

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.

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?

Page 6, Line 3-4: The SP2 scattering channel was not saturated for the 500 nm and 1000 nm PSLs? 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.

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

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experiment also be included in this table?

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.

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.

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

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.

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

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.

Page 11, Line 29-30: What is the reason for focusing on the time delay analysis data

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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?

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?

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.

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.

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.

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

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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.

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.

Technical Corrections: Page 2, Line 3: remove “the” from “. . . play a vital role in the climate change. . .”

Page 2, Line 5: remove “their” from “its their”

Page 2, Line 11: remove “, evidently”

Page 2, Line 19: remove “much”

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”

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

Page 4, Line 20: Remove “generally”

Page 5, Line 32: change “fitted” to “fit”

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

Page 14, Line 5: Change “open” to “laboratory”

Page 14, Line 13: Remove “urgently”

Page 14, Line 26: Remove “obviously”

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

Table 1: Change “C/S ratio” to “S/C ratio”

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

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.

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.

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.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2017-326, 2017.