### **Response to comments of anonymous referees #1**

Review of the manuscript titled: "Physical and chemical properties of black carbon and organic matter from different sources using aerodynamic aerosol classification" by Dawei Hu et al.

The paper describes laboratory measurements of the physical properties of black carbon particles emitted from different sources. The paper is overall well written (with some relatively minor issues as discussed later), and the approach seems quite comprehensive and, for the most part, sound. The results provided by this study are important for the community and I would like to see them published. The paper requires some significant but relatively straightforward revisions, after which, the paper can be most probably published.

We thank the reviewer for the positive comments on our manuscript. In the revised version, we have addressed the comments listed below.

### **General comments**

• A good part of the introduction focuses on optical properties and refractive indices, but then at the end of it, the authors mention that the optical properties are not the subject of the current paper. I would suggest refocusing the introduction on the topic of the paper.

The introduction is rephrased in the revised manuscript (line 63-161 on page 3-7). More discussions regarding the physical properties of particles are added in line 80-108 on page 4 and 5.

"Although BC and BrC are very important for climate, they are poorly represented in atmospheric models (Zuidema et al., 2016). This is in part due to the complex microphysical properties of BC and the lack of accurate refractive index (RI) descriptions for both BC and BrC (Liu et al., 2020). Fresh soot particles often exist in the form of aggregates composed of primary spherules with an irregular and highly fractal geometry (Xiong and Friedlander, 2001;Wentzel et al., 2003). The morphology of these aggregates change markedly during the atmospheric aging process, influencing the corresponding particle size and optical properties (Zeng et al., 2019;Zhang et al., 2008). For example, after condensation of gaseous species such as sulfuric acid or water (under high relative humidity (RH) environments) on soot particles, or coagulation with the pre-existing particles, soot particles can experience restructuring and the shape of the soot particles becomes more similar to a spherical particle (Zhang et al., 2008). The morphology of BC particles can be measured directly by using Scanning Electron Microscopy (SEM) or Transmission Electron Microscopy (TEM) (Fu et al., 2006; Chen et al., 2018; Ellis et al., 2016). However, the SEM/TEM approach only provides particle shape information in two dimensions and do not provide real time characterisation. Alternatively, the particle morphology can be determined by measuring its size and mass with different techniques (Chen et al., 2018;DeCarlo et al., 2004). A conventional approach is to classify particles (generally using a differential mobility analyser, DMA, to select monodisperse particles on their mobility size) and then measure particle mass using a particle mass analyser (Zhang et al., 2008;Park et al., 2003;Park et al., 2004a;Park et al., 2004b;Chen et al., 2018) (Wu et al., 2019). From the resulting information about particle mass for different particle mobility sizes, the dynamic shape factor ( $\chi$ , defined as the ratio of the drag force on the particle divided by the drag force on the particle's volume equivalent sphere) and fractal dimensions ( $D_f$ ) can be retrieved (DeCarlo et al., 2004). Mobility-mass fractal dimension (D<sub>fm</sub>) has been reported over a wide range of 2.2-2.8 for diesel exhaust particles (Park et al., 2004b).  $D_{fm}$  has been reported as higher than the  $D_{f_1}$  - defined as the scaling exponents between the radius of gyration of an aggregate and the radius of primary spherules composing the aggregate - but the two are not always directly equivalent, particularly in the transition regime (Sorensen, 2011)."

• The excessive use of emphatic words in the abstract/introductions such as "pioneering", "authoritative", "novel", etc. detracts from the undoubted value of the work. I would suggest removing these terms that are just irritating and add nothing to the paper.

#### These terms are removed in the revised manuscript.

• How do multiple charges affect the mass measurements provided by the CPMA and how is that accounted for?

In this study, as the particles from each source (either engine, wood combustion or flame burner) will have a single effective density for a given size, the AAC will deliver particles of only a single physical size. Therefore there will be no larger particles available to be multiply charged prior to sizing by the CMPA.

Results are reported without uncertainties, making comparisons, and the understanding of the significance of the results difficult. Please estimate potential uncertainty bounds (both statical as well as biases) for all the quantities reported or calculated including chemical, morphological, or other physical quantities such as densities, fit slopes, shape factors, etc.

The uncertainties are calculated and added for the data regarding the shape factor of BC (Table 1 and Figure 7, The error bar in Fig. 7(a) and Fig. 7(b) is too small to see clearly), material density of organics (Table 1), fit slopes for the SP2 incandescence signal calibration (Figure 4, the uncertainties refer to fitting precision based on the standard error in the regressed slope). The noise to signal ratio (<0.005) is also provided of the organic mass spectrum. In addition, all these above uncertainties are now quoted in the main text (e.g., in Sect. 3.4 for the reported dynamic shape factors and densities, line 710 for the noise to signal ratio, and lines 765-766 for the SP2 incandescence signal correction factor).

• Some grammatical and tense consistency checks would be advisable (limited examples in the specific comments next).

## The grammar and tense have been checked for consistency throughout the manuscript.

 References are somewhat scarce and myopic, neglecting some important related work especially on the AAC/CPMA/DMA use, BC morphology, and SP2 signal interpretation. I did not provide too many specific examples below just because there is a lot of work out there that seems very relevant to this study.

More related references (listed below) have been added and discussed in the introduction of the revised manuscript.

Chen, C., Enekwizu, O. Y., Fan, X., Dobrzanski, C. D., Ivanova, E. V., Ma, Y., et al., 2018. Single Parameter for Predicting the Morphology of Atmospheric Black Carbon. Environmental Science & Technology. 52, 14169-14179, <u>http://doi.org/10.1021/acs.est.8b04201</u>. DeCarlo, P. F., Slowik, J. G., Worsnop, D. R., Davidovits, P., and Jimenez, J. L., 2004. Particle Morphology and Density Characterization by Combined Mobility and Aerodynamic Diameter Measurements. Part 1: Theory. Aerosol Science and Technology. 38, 1185-1205, <u>http://doi.org/10.1080/027868290903907</u>.

Ellis, A., Edwards, R., Saunders, M., Chakrabarty, R. K., Subramanian, R., Timms, N. E., et al., 2016. Individual particle morphology, coatings, and impurities of black carbon aerosols in Antarctic ice and tropical rainfall. Geophysical Research Letters. 43, 11,875-811,883, <u>http://doi.org/10.1002/2016gl071042</u>.

Fu, F., Xu, L., Ye, W., Chen, Y., Jiang, M., and Xu, X., 2006. Morphology of black carbon aerosols and ubiquity of 50-nanometer black carbon aerosols in the atmosphere. China Particuology. 4, 323-326, <u>http://doi.org/10.1016/S1672-2515(07)60283-8</u>.

Moteki, N., and Kondo, Y., 2007. Effects of Mixing State on Black Carbon Measurements by Laser-Induced Incandescence. Aerosol Science and Technology. 41, 398-417, <u>http://doi.org/10.1080/02786820701199728</u>.

Park, K., Cao, F., Kittelson, D. B., and McMurry, P. H., 2003. Relationship between Particle Mass and Mobility for Diesel Exhaust Particles. Environmental Science & Technology. 37, 577-583, <u>http://doi.org/10.1021/es025960v</u>.

Park, K., Kittelson, D., Zachariah, M., and McMurry, P., 2004a. Measurement of Inherent Material Density of Nanoparticle Agglomerates. Journal of Nanoparticle Research. 6, 267-272, <u>http://doi.org/10.1023/B:NANO.0000034657.71309.e6</u>.

Park, K., Kittelson, D. B., and McMurry, P. H., 2004b. Structural Properties of Diesel Exhaust Particles Measured by Transmission Electron Microscopy (TEM): Relationships to Particle Mass and Mobility. Aerosol Science and Technology. 38, 881-889, <u>http://doi.org/10.1080/027868290505189</u>.

Sedlacek, A. J., Onasch, T. B., Nichman, L., Lewis, E. R., Davidovits, P., Freedman, A., et al., 2018. Formation of refractory black carbon by SP2-induced charring of organic aerosol. Aerosol Science and Technology. 52, 1345-1350, http://doi.org/10.1080/02786826.2018.1531107. Sorensen, C. M., 2011. The Mobility of Fractal Aggregates: A Review. Aerosol Science and Technology. 45, 765-779, <u>http://doi.org/10.1080/02786826.2011.560909</u>.

Tavakoli, F., Symonds, J. P. R., and Olfert, J. S., 2014. Generation of a Monodisperse Size-Classified Aerosol Independent of Particle Charge. Aerosol Science and Technology. 48, i-iv, <u>http://doi.org/10.1080/02786826.2013.877121</u>.

Wentzel, M., Gorzawski, H., Naumann, K. H., Saathoff, H., and Weinbruch, S., 2003. Transmission electron microscopical and aerosol dynamical characterization of soot aerosols. Journal of Aerosol Science. 34, 1347-1370, <u>http://doi.org/10.1016/S0021-8502(03)00360-4</u>.

Wu, Y., Xia, Y., Huang, R., Deng, Z., Tian, P., Xia, X., et al., 2019. A study of the morphology and effective density of externally mixed black carbon aerosols in ambient air using a size-resolved single-particle soot photometer (SP2). Atmos. Meas. Tech. 12, 4347-4359, <u>http://doi.org/10.5194/amt-12-4347-2019</u>.

Xiong, C., and Friedlander, S. K., 2001. Morphological properties of atmospheric aerosol aggregates. Proceedings of the National Academy of Sciences. 98, 11851-11856, <u>http://doi.org/10.1073/pnas.211376098</u>.

Zeng, C., Liu, C., Li, J., Zhu, B., Yin, Y., and Wang, Y., 2019. Optical Properties and Radiative Forcing of Aged BC due to Hygroscopic Growth: Effects of the Aggregate Structure. Journal of Geophysical Research: Atmospheres. 124, 4620-4633, <u>http://doi.org/10.1029/2018jd029809</u>.

Zhang, R., Khalizov, A. F., Pagels, J., Zhang, D., Xue, H., and McMurry, P. H., 2008. Variability in morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing. Proceedings of the National Academy of Sciences. 105, 10291, <u>http://doi.org/10.1073/pnas.0804860105</u>.

# **Specific comments**

Line 44: "This implies" or "This suggests", how certain are the authors about the following statement?

### "This implies" is modified to "This suggests"

Line 51: Please provide uncertainty bounds for these values, otherwise it is hard to understand if the later statement (on line 55) on the difference from the 0.75 value might be justified; in other words, is the difference significant?

The uncertainty is analysed and discussed in the revised manuscript.

The sentence is modified to: "A correction factor is defined as the ratio of the incandescence signal from an alternative BC source to that from the Aquadag standard, and took values of  $0.821 \pm 0.002$  (or  $0.794 \pm 0.005$ ),  $0.879 \pm 0.003$  and  $0.843 \pm 0.028$  to  $0.913 \pm 0.009$  for the BC particles emitted from the diesel engine running under hot (or cold idle) conditions, the flame burner and wood combustion, respectively."

Line 50-54 on page 2 and 3.

Line 75: The statement that the absorption coefficient for BC is wavelength-independent is incorrect, the typical dependence, as extensively reported in the literature, is often expressed as a power law with an exponent of about -1 (which is still a strong wavelength dependence, although weaker than that of brown carbon). What is often assumed (but probably also not always true) is that the imaginary part of the index of refraction is wavelength-independent (or at least not very strongly dependent). BrC also has an absorption that is wavelength dependent dependent just with an exponent that is significantly larger, in absolute value, than that of BC.

The sentence is modified to: "Typically, the light absorption coefficient for BC and BrC is wavelength dependent over the visible spectrum, with BrC exhibiting a stronger wavelength dependence characterised by increasing absorption at progressively shorter visible wavelengths (Kirchstetter et al., 2004;Corbin et al., 2019;Voliotis et al., 2017)."

Line 76-79 on Page 3 and 4.

Line 95: This is an interesting approach but it is hardly pioneering, I would call this incremental in a very positive sense (see, for example, the work by the Olfert's group, or others). I suggest removing this exaggerated adjective and point to existing literature. Same in line 99.

The term "pioneering" is removed.

The sentence is revised to: "To address the issues mentioned above, the Soot Aerodynamic Size Selection for Optical properties (SASSO) project utilised the Aerodynamic Aerosol Classifier (AAC) to classify particles according to aerodynamic diameter for size and mass distribution measurements and optical evaluation (Tavakoli et al., 2014). Specifically, SASSO has used the AAC size selection of emissions from wood burning, diesel combustion and secondary organic aerosol (SOA) formation, prior to optical measurements using cavity ring-down and photoacoustic spectroscopy with the EXtinction, SCattering and Absorption of Light for AirBorne Aerosol Research (EXSCALABAR) instrumentation, custom-built by the Met Office (Cotterell et al., 2020;Cotterell et al., 2019;Davies et al., 2018)."

## Line 133-141 on page 6.

Line 97: How do the authors determine themselves that the method is "authoritative"? That, if true, should be a judgment left to the community.

The word "authoritative" is deleted.

Line 111: In what way does the SP2 provide information about the morphology? The information is likely limited and subject to large uncertainties. Several papers have been published on the topic, some in contrast with others.

The SP2 may provide some morphology by analysing the relative peak position between scattering and incandescence signal, however this technique is not quantitative which is subject to considerable uncertainties, and literatures reported contrasting results. We therefore use size/mass or TEM measurements to obtain the morphology rather than using the SP2 itself.

The "morphology" term is removed and the sentence is revised to:

"Important additional considerations in the retrieval of refractive indices from optical spectroscopy data are the aerosol morphology (described above) and mixing state. The mixing state can be probed using the Single Particle Soot Photometer (SP2), which can measure the refractory BC (rBC) mass content and optical size of individual particles. However the SP2 needs an empirical calibration to retrieve the rBC mass from the incandescence signal (Laborde et al., 2012a). The conventional method to calibrate the incandescence channel of SP2 is using size selected Aquadag standards (Acheson Inc. USA) and then correcting to a calibration representative of ambient rBC by a constant factor of 0.75 (Laborde et al., 2012b). However, few experiments since have independently verified this across various soot types."

Line 123-132 on page 5 and 6.

Line 116: Consider rewording "which makes the complexity of the calibration methods" to "which makes the calibration methods complex" or "challenging"

This is revised.

Line 118: Change "corrected" to "correcting"

This is revised.

Line 135 "to to" -> "to"

This is revised.

Line 146: The AAC select aerosol by aerodynamic size; so, aerosol particles passing through it are indeed monodisperse in terms of aerodynamic size, but that does not mean that the output distribution is mondisperse in every size measure; for example, particles of the same mass (and therefore mass-equivalent diameter) could have very different aerodynamic size depending on their morphology. So, the term monodisperse here is ambiguous. And it all depends on the property one wants to measure (for example, absorption mostly depends on mass).

This sentence is modified to: "The AAC (Cambustion Ltd, Cambridge, UK) is used to select aerosols within a narrow range of aerodynamic diameters and does not suffer from the issue of multiple charges that affects selection using instruments such as the CPMA and DMA."

Line 180-182 on page 8.

We modified the term of "monodisperse particles" associated with AAC to "particles classified within a narrow range of aerodynamic diameter" throughout the whole manuscript.

Line 200: How well does an optical size measurement calibrated with PSLs perform on fractal-like black carbon particles? Is the size an optical equivalent to a spherical PSL particle? That should be mentioned as the meaning of "size" for a fractal-like particle is always quite ambiguous (see the previous comment as well).

## A sentence is added for further clarification:

"The particle size can be determined by detecting the laser signal scattered by particles, with the scattering intensity maximum related to the optical particle diameter through a calibration using polystyrene latex spheres. The optical size of BC-containing particles is determined by matching the measured scattering signal with calculations from light scattering calculations assuming a core-shell structure (core-shell Mie theory) (Moteki and Kondo, 2007)."

### Line 229-234 on page 10.

Line 217: Change verb in the sentence "The instrument operation and data analysis of HR-AMS has been..." to "The instrument operation and data analysis of HR-AMS have been..." for number consistency.

### This is revised.

Section 2.1.5: The CPMA, using an electric field, also suffers from the issue of multiple charges as in the case of the DMA, this should be mentioned. Also, what charge neutralizer was used for the CPMA should be mentioned for consistency with the following description of the SMPS.

The following sentence is added for further clarification.

"As the CPMA uses an electrical classification method to select particles, it also suffers from an issue of multiple charging similar to a DMA, and it has problems associated with a fraction of the uncharged particles that are also transmitted, particularly at the lower rotation speeds. In this study, an electrical ioniser (MSP Corp., USA) was used for wood combustion experiments and a <sup>90</sup>Sr radioactive ioniser was used for chamber experiments to neutralize particles before they were sampled by the CPMA."

Line 269-274 on page 12.

Line 259: It would be good to provide a reason behind the choice of the denuder temperature set point.

For the purpose of this study, we wanted to remove as much of the organic coating material from the combustion-generated particles as possible and 180 °C is the maximum temperature for our self-built thermal denuder.

The following sentence is added for further clarification.

"In this study, the purpose of the TD is to remove as much of the organic coatings from the combustion-generated particles as possible, rather than probing the volatility properties of the coating. Therefore, all heating zones of the TD were set to their maximum temperature of 180 °C. This upper temperature is lower than that achieved by other commercial TD units and minimises the risks of charring."

Line 304-309 on page 13.

Line 285: Suggest changing "can be" to "to be"

This is revised.

Line 285-286: Do the authors have a more quantitative measure of the aerosol loss rate?

Yes, the wall losses of particles inside the Manchester aerosol chamber was investigated and the corresponding results is under review in AMT (Shao et al., 2021). Briefly, a series of experiments were conducted to investigate the size-resolved particle lifetimes under various humidity and mixing conditions using ammonium sulfate particles, which was introduced to the chamber and left in the dark at the desired RH and temperature conditions for  $\geq$ 4 hours. The mean number and mass wall loss rates were estimated as  $9.17 \pm 1.3$  and  $8.16 \pm 1.5 \times 10^{-5}$  s<sup>-1</sup>, respectively. More details can be found in section 3.5 in Shao et al. (2021).

The following sentence is added in the revised manuscript.

"The relatively large volume of the chamber allows the dilute sample to be held for several hours without significant aerosol removal from wall losses, allowing the study of particles introduced directly or formed within the chamber over a period of several hours. The mean

number and mass wall loss rates of particles inside the chamber were estimated as  $9.17 \pm 1.3$ and  $8.16 \pm 1.5 \times 10^{-5}$  s<sup>-1</sup>, respectively (Shao et al., 2021)."

Line 335-339 on page 15.

Reference:

Shao, Y., Wang, Y., Du, M., Voliotis, A., Alfarra, M. R., Turner, S. F., et al., 2021. Characterisation of the Manchester Aerosol Chamber facility. Atmos. Meas. Tech. Discuss. 2021, 1-50.

Line 329: How many iterations does the process typically take?

We used the Igor Pro 'Findroots' command, which uses the standard Brent's method for solving functions. It does not report the number of iterations.

More information is added in line 381-383 on page 17:

"The equations were solved iteratively using a standard Brent's method solver ('findroots' command, Igor Pro version 6.36, Wavemetrics)."

Line 366: Something awkward about this sentence. Maybe "igniting" should be "ignited" or "ignites"?

"igniting" is revised to "ignites".

Line 387: Remove "in" or "during"

# "in" is removed

Line 476: Just a comment: interestingly, these results seem similar to what was reported by Bhandari, et al. Scientific Reports 9(1): 11824 (2019)

Yes, our results are similar to those reported by Bhandari, et al. (2019). This reference is cited in the revised manuscript.

Line 482: I am confused by the potential explanation (2), and maybe I missed something, but I thought at least in some of the experiments that the particles were minimally coated, so how would the size be dominated by organic coatings, also in those cases?

More information is added in line 680-687 on page 29.

"There are two possibilities: (1) The BC cores retained their structure throughout humidity cycling process; or (2) As shown in Figure 8, the coatings on BC particles are very thick and therefore dominate the particle, rendering our size measurement approach insensitive to any changes in BC core size from restructuring. The size of the bare BC particles is around 68 nm, but after coating by SOA, the size of the coated BC particles reaches up to around 300 nm. Due to the very large coating thicknesses of the coated BC particles, even if the BC cores were restructured during the humidity cycling process, any changes were not reflected in the overall particle size as this was dominated by the contribution from the coating organics."

Line 490: This is a very small diameter. How large were the monomers in these BC particles, and how many monomers typically in an aggregate? Were these particles made of only a very few monomers?

As shown the TEM images in Fig. 7(c), the primary spherule size of the BC particles from the diesel engine is around 10-15 nm. However, we do not have micrographs for these specific experiments, so cannot report on the precise numbers of monomers in this case.

Line 501: Maybe replace "improve" with "improving"?

### This is revised.

Figure 6: Especially for Aquadag (but it might be slightly visible also in some of the other BC types), there seems to be a slight negative curvature in the graphs (especially visible in the center and right graphs). What is the reason for such a change in slope? One could study these changes of the slope by graphing residuals plots. I believe Aquadag comes already compacted; is it possible that the compacted morphology "shields" the aggregate lowering the incandescence signal at higher masses with respect to what might be expected for not compacted BC particles of the same mass, resulting in the negative curvature?

We believe the deviation from linear to only be very slight and small compared to the variations in slopes, which is the key result being presented here. While the effect described by the reviewer may be plausible, we should point out that there may be a competing effect if resonances occur within spherical particles. But whichever way, it is generally assumed that the interactions between the BC and the incident light occur within the Rayleigh regime (Moteki and Kondo, 2007), where either effect should be very minor. Regardless, we do not believe that we can conclude anything firm based on this data, so consider this outside of the paper's scope.

#### Reference:

Moteki, N., and Kondo, Y., 2007. Effects of Mixing State on Black Carbon Measurements by Laser-Induced Incandescence. Aerosol Science and Technology. 41, 398-417, http://doi.org/10.1080/02786820701199728.

Lines 510 to 526: These are very interesting results, but uncertainty bounds should be reported to understand how significant these differences are. How the uncertainties (both statistical and systematic) are estimated, should also be carefully described.

The uncertainties are calculated and added and discussed in the revised manuscript.

"In this study, the incandescence signal of the SP2 was measured for BC particles from catalytically stripped diesel engine exhaust emissions, an inverted flame burner, and controlled flaming wood combustion, respectively, and compared with that measured from an Aquadag standard. The uncertainties here refer to precision of the fitted parameters reported by the Igor Pro fitting algorithm, based on analysis of residual data. As shown in Fig. 4, for the BC particles emitted from the diesel engine under hot engine and cold idle conditions (Fig. 4(a)), the slopes of the incandescence signal with BC mass are  $0.821 \pm 0.002$  and  $0.794 \pm 0.005$  times of that measured from the Aquadag standard, respectively. Note that while some deviation from a perfect linear response is noted, this is small compared to the variation in slopes, so represents a minor source of uncertainty in comparison. These correction factors are 9.4% and 5.6% different with the common value of 0.75 (with the uncertainty less than 5%) recommended by Laborde et al. (2012b) when deriving the mass concentration of BC emitted from diesel engines. For the BC particles generated from the flame burner (Fig. 4(b)), the correction factor is  $0.879 \pm 0.003$ . Meanwhile, for the BC particles emitted from the

flaming phase during the combustion of Scots pine, Poplar, Giant Redwood or Western red cedar, the correction factors are  $0.913 \pm 0.009$ ,  $0.906 \pm 0.014$ ,  $0.889 \pm 0.027$  or  $0.843 \pm 0.028$ , respectively. We stress that, for the SP2 calibrations here from wood combustion emissions, the BC particles were not treated with a catalytic stripper before sampling by the SP2. While coating materials may char under 1064 nm to produce refractory black carbon and therefore cause overestimates in the incandescence signal (Sedlacek et al., 2018), as shown in Fig. 2(c), the BC particles generated at the beginning of the flaming phase contained almost no organic species, with  $r_{OA}$  values less than 0.05. Even if this OC were to be converted to EC with 100% efficiency (which we consider to be highly unlikely), this would represent a very small error.

The differences in the correction factors derived in this study with the default value of 0.75 are 9.4% (5.6%), 17.2% and 12.4-21.7% for the BC particles emitted from engine with hot engine (or cold idle) condition, flame burner and wood combustion, respectively. We recommend that future studies utilizing the SP2 for rBC mass concentration measurements use SP2 calibrations with the same type of BC as that to be studied."

Line 554-581 on page 24 and 25.

Lines 516-521: This means that some organics still coat the BC particles, even if in a small amount, correct? Is it possible that some of this organic would char and generate an incandescence signal like that of BC? See, for example, Sedlacek, et al. Atmos. Chem. Phys. 18: 11289-11301 (2018).

The reviewer is correct in saying that the BC particles produced from the flaming wood combustion may have some organic components, particularly given that the combustion-generated particles were not treated with a catalytic stripper before sampling by the SP2. Based on the study by Sedlacek et al. (2018), some of coating materials may char under 1064 nm to produce the refractory black carbon and then overestimate the incandescence signal. However due to experimental limitations, we were unable to confirm the presence of this phenomenon. In this study, as the measured organic mass fraction in the BC particles is less than 0.05, the charring effect would be very limited.

A sentence is added to discuss the charring effect in line 570-576 on page 25.

"We stress that, for the SP2 calibrations here from wood combustion emissions, the BC particles were not treated with a catalytic stripper before sampling by the SP2. While coating

materials may char under 1064 nm to produce refractory black carbon and therefore cause overestimates in the incandescence signal (Sedlacek et al., 2018), as shown in Fig. 2(c), the BC particles generated at the beginning of the flaming phase contained almost no organic species, with  $r_{OA}$  values less than 0.05. Even if this OC were to be converted to EC with 100% efficiency (which we consider to be highly unlikely), this would represent a very small error."

Section 3.2: As mentioned in the general comments, here (as in other places in the paper) a comparison is difficult without having a good estimate of how certain these reported values might be.

The uncertainties are calculated and added in Table 1, Figure 7 and the manuscript.

Lines 644-645: What does it means that "the peaks are most dominated in the smouldering phase"? Do they mean "are most dominant in the smouldering phase" or something else? Also, check tense consistency with just a couple of lines earlier

Yes, it should be "the peaks are most dominant in the smouldering phase". This is modified in the revised manuscript.

The tense consistency is checked in the revised manuscript.

Line 653: "in" in front or "contrast".

This is revised.

Lines 669 – 671: "clear difference... was" or "clear differences ... were" but not "clear difference... were"

This is revised.