This manuscript pulls from the laser-induced incandescence (LII) literature and expands its application to optical-acoustic phenomena, with a specific focus on the effect of agglomeration and characteristics cooling times. It is proposed that such a model may be useful in accounting or correcting for particle composition and morphology. The manuscript does not adequately reference the existing LII literature on the topic, which must be improved (both in the introduction and but also in terms of recurring references to the literature throughout the text). However, the application of a simple version of existing LII models to optical-acoustic measurements remains interesting. The manuscript is reasonably well-written overall.

Thus, while the manuscript requires some reframing and improvements in references to the existing literature and could benefit from further model validation (comparison of temporal signals), the manuscript could be a good addition to the literature and should be considered for publication following several changes.

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(1) The review of the LII modeling literature is seemingly random. Given the perceived focus on models in the literature, a key omission was the review and comparison of LII models by Michelsen et al. in 2007. This would cover many of the earlier models, while covering some missed by the authors. There are also key manuscripts that discuss conduction that are likely relevant, including a key manuscript by Filippov et al. (2000) and several other manuscripts by Liu et al. and Daun (2010). Some of these manuscript also mention the effect of shielding, which results in an uneven distribution of temperature in the aggregate. This reviewer would encourage the authors to re-review the corresponding literature. Liu et al. (Appl. Phys. B, 2006b) is mentioned later but seems like an omission in the literature review earlier in the manuscript. Given these omissions, the choice to cite a manuscript for 3D LII that is marginally relevant is poor, even if such a study is interesting and robust.

Michelsen, H.A., Liu, F., Kock, B.F., Bladh, H., Boïarciuc, A., Charwath, M., Dreier, T., Hadef, R., Hofmann, M., Reimann, J. and Will, S., 2007. Modeling laser-induced incandescence of soot: a summary and comparison of LII models. Applied physics B, 87(3), pp.503-521.

Filippov, A.V., Zurita, M. and Rosner, D.E., 2000. Fractal-like aggregates: relation between morphology and physical properties. Journal of colloid and interface science, 229(1), pp.261-273.

Liu, F., Smallwood, G.J. and Snelling, D.R., 2005. Effects of primary particle diameter and aggregate size distribution on the temperature of soot particles heated by pulsed lasers. Journal of Quantitative Spectroscopy and Radiative Transfer, 93(1-3), pp.301-312.

Liu, F., Yang, M., Hill, F.A., Snelling, D.R. and Smallwood, G.J., 2006a. Influence of polydisperse distributions of both primary particle and aggregate size on soot temperature in low-fluence LII. Applied Physics B, 83(3), pp.383-395.

Liu, F., Daun, K.J., Snelling, D.R. and Smallwood, G.J., 2006b. Heat conduction from a spherical nano-particle: status of modeling heat conduction in laser-induced incandescence. Applied physics B, 83(3), pp.355-382.

Liu, F. and Smallwood, G., 2008. Study of heat conduction between fractal aggregates and the surrounding gas in the transition regime using the DSMC method. In 40th Thermophysics Conference (p. 3917).

Daun, K.J., 2010. Effect of selective accommodation on soot aggregate shielding in time-resolved laser-induced incandescence experiments.

We would honestly like to express our gratitude to the reviewer for providing this historically consistent story telling of improving the description of agglomerate particle heat terms. We have to admit that we only used the most relevant of these references in our manuscript, which somehow did not provide the full scope of the relevant work. In the revised version, we have included the additional references provided both in the introduction and in the methodology section as follows:

Introduction: “Dissipation of energy from BC particles has been analysed in detail for LII sensors. Filippov (2000) were the first to propose a model for particle sizing based on energy dissipation and Michelsen (2003), Michelsen et al. (2007) and Michelsen et al. (2015) later elaborated and demonstrated a model that describes several dissipation mechanisms for BC particles. Hadef et al. (2010) presented a simpler version with the most relevant mechanisms for LII sensors. Snelling et al. (2004) used a similar model and performed an experimental evaluation to fine-tune the heat conduction term. The model was further extended by a series of additional works by the same research group that demonstrated application of that model on specific LII experiments (Liu et al. 2005a, Liu et al. 2006a) that led to a more refined heat dissipation theory by spherical particles (Liu et al. 2006b). Starke et al. (2003) developed an algorithm that can calculate the average particle size based on the delay of the LII signal. In addition, Meyer et al. (2016) showed that 3D images of BC particles can be constructed from LII signal analysis. Willems et al. (2019) developed a model that combine signals from LII and OA sensors. However, the dissipation mechanisms are only used for the LII sensor, and the OA signal is used as an independent additional input. A theoretical model that applies similar LII energy dissipation mechanisms of BC particles for OA sensors is still missing.”

Methodology: “We use an energy balance model to simulate the light-to-heat processes for BC particles, following the principles of the models developed for LII applications. In particular, the mathematical formulation for absorption, conduction, sublimation, and radiation follows the description by Mikkelsen et al. (2003). A compact version of this model is described by Hadef et al. (2010), which is used as the starting point for the current analysis”

(2) The authors should consider rewriting the model description. The reliance on Hadef et al. as a starting point both seems both arbitrary and inefficient. The model resembles others in the LII literature in key ways (e.g., Liu et al., 2006), with the two key terms (conduction and absorption) not resembling that in Hadef et al. It would seem that the better way to introduce the model is to state how all of the terms are being treated explicitly before stating that remaining terms are treated the same as or have the same parameters as Hadef et al. The authors should consider the broader set of models presented in Michelsen et al. (2005, 2015) and newer models when presenting their own model.

Indeed, our reference to Hadef’s work should be done more sparingly, as this is not the original source of the modelling theory but rather only a compact implementation of this. We have now made this clear in our manuscript as suggested by the reviewers. Moreover, we have refrained from making references to that model throughout the manuscript, and instead, we make reference to original sources, as required.

For example for absorption the updated model description is: “Typically (Mikkelsen et al. 2003 and references therein) particle absorption efficiency (Q_{abs}) is calculated using the particle size, the light wavelength, and an experimental absorption function E_m as inputs”

(3) Why do the authors place so much emphasis on the agglomeration for conduction but still apply Mie theory for modeling the absorption? Why is Mie theory applied to the individual monomers and then RDG-FA applied to the agglomerates? In regards to the validity of RDG-FA, consider citing work by Sorensen, Liu, Yon, and co-workers (see below), who provide overviews of the approximate validity of RDG-FA as well as when errors can be incurred. Of particular note, Liu et al. (2010) looks at the effect of inaccuracies in RDG-FA on LII temperature decays, with some interplay with conduction.

Liu, F. and Smallwood, G.J., 2010. Effect of aggregation on the absorption cross-section of fractal soot aggregates and its impact on LII modelling. Journal of Quantitative Spectroscopy and Radiative Transfer, 111(2), pp.302-308.

Yon, J., Liu, F., Bescond, A., Caumont-Prim, C., Rozé, C., Ouf, F.X. and Coppalle, A., 2014. Effects of multiple scattering on radiative properties of soot fractal aggregates. *Journal of Quantitative Spectroscopy and Radiative Transfer*, *133*, pp.374-381.

Sorensen, C.M., Yon, J., Liu, F., Maughan, J., Heinson, W.R. and Berg, M.J., 2018. Light scattering and absorption by fractal aggregates including soot. Journal of Quantitative Spectroscopy and Radiative Transfer, 217, pp.459-473.

We have further elaborated on the method used for estimating particle absorption and introduced the references suggested by the reviewer, as they – again – better reflect original source of reference information. We now believe that the length of analysis between absorption and conduction is more balanced. We also present the limitations of our approach in, making clear this is not critical for OA particle sizing, unlike LII. We believe it is clear that energy dissipation – mostly conduction in our case – is much more critical for our model than the absolute absorption calculation. The new section is the following:

"Agglomerate particle absorption is estimated using the RDG-FA method (Sorensen et al. 2018; Liu et al. 2010, Romshoo et al. 2022). For applying this method, we calculate the absorption coefficient of the primary particles that form the agglomerate using the Mie theory for spherical particles (Hinds, 1999), the refractive index suggested by Bond & Bergstrom (2006) which is equal to 1.9 – 0.75i, and a handy Mie Simulator GUI (2025). This simulator outputs the absorption efficiency of the primary particle as a function of light wavelength. To obtain a full picture of the absorption coefficient dependence on various primary particle sizes, we run the simulator over a wide size range of 10 to 1000 nm. For validation, the new absorption calculation method was tested with the conditions of Hadef et al. (2010) and produced similar results for the same input. The advantage of this rather simple proposed method is that it can fine-tuned for particles of different refractive indices, without the need for conducting new experiments. However, Liu et al. (2010) and Bond et al. (2006) have demonstrated that this method may underestimate absorption by up to 30%. Unlike LII applications, where absorption is critical for particle peak temperature, the absolute absorption term is not crucial for our model, which focuses on the energy dissipation terms to characterize particles.

The particle heat conduction term has been described by different models, as outlined by Liu et al. (2006). The Boundary-Sphere Method (BSM) is used here as a good compromise between accuracy and complexity. More elaborate analysis shows that shielding of external boundary spherules is responsible for 3-4% deviation in heat dissipation terms (Daun, 2010), which is not considered in our model. Key to the BSM approach is the particle Knudsen number (*Kn*). The volume around the particle is split into two regions, one close to the particle where heat conduction from the particle to the boundary sphere takes place in the free-molecular regime ($Kn > 1$), and an outer region where heat from the boundary sphere to the surrounding air dissipated in the continuum regime (*Kn* < 1). The boundary sphere is assumed to have a uniform temperature (T_δ) and its radius is equal to the radius of the particle plus a parameter *δ*."

(4) "The diameter of gyration is a proxy of the wetted surface of the particle, which is relevant for heat conduction." – Minor point, but can the authors provide a citation supporting this statement.

Indeed. We improved the expression to : 'The diameter of gyration is a good proxy of the equivalent heat conduction radius (Liu 2008)".

(5) "The thermal relaxation time of a spherical particle can be calculated on the basis of Eq. (1) as shown in Starke et al. (2003)." – Better references for this may be the original papers by Eckbreth and Melton.

Eckbreth, A.C., 1977. Effects of laser-modulated particulate incandescence on Raman scattering diagnostics. *Journal of Applied Physics*, *48*(11), pp.4473-4479.

Melton, L.A., 1984. Soot diagnostics based on laser heating. Applied optics, 23(13), pp.2201-2208.

Thank you for your comment. We added the two references at the specified section.

(6) Sec. 2.4. This kind of observation has been observed previously, relating the cooling time to the Sauter mean diameter of the particles (Liu et al., 2005, 2006). This was also stated in the review by Schulz et al. (2006). This has been affirmed in later literature when examining uncertainties from experimental data (Daun et al., 2007; Sipkens et al., 2014). It is thus unsurprising and not particularly novel that the data collapsed when a Sauter mean diameter (or surface-to-volume ratio) is considered, though the extension to optical-acoustic measurements is notable. The authors should acknowledge the corresponding literature. Polydispersity also disrupts this trend, causing some scatter that the authors should acknowledge and was observed in the above studies.

Liu, F., Snelling, D.R. and Smallwood, G.J., 2005, January. Numerical study of temperature and incandescence intensity of nanosecond pulsed-laser heated soot particles at high pressures. In ASME International Mechanical Engineering Congress and Exposition (Vol. 42215, pp. 355-364).

Schulz, C., Kock, B.F., Hofmann, M., Michelsen, H., Will, S., Bougie, B., Suntz, R. and Smallwood, G., 2006. Laser-induced incandescence: recent trends and current questions. Applied Physics B, 83(3), pp.333-354.

Daun, K.J., Stagg, B.J., Liu, F., Smallwood, G.J. and Snelling, D.R., 2007. Determining aerosol particle size distributions using time-resolved laser-induced incandescence. *Applied Physics B*, *87*(2), pp.363-372.

Sipkens, T.A., Mansmann, R., Daun, K.J., Petermann, N., Titantah, J.T., Karttunen, M., Wiggers, H., Dreier, T. and Schulz, C., 2014. In situ nanoparticle size measurements of gas-borne silicon nanoparticles by time-resolved laser-induced incandescence. *Applied Physics B*, *116*(3), pp.623-636.

This is another good point that we were unaware of and – again – thank the reviewer for the suggestion. We have included the relevant discussion and we are further relieved that one of our intermediate findings is confirmed. We have added the following:

"A possible explanation for this is that the volume of the particle is proportional to the amount of energy absorbed, while the surface calculated with the diameter of the agglomerate particle is a good proxy for the heat equivalent diameter of the particle. Thus, the V/S ratio is a good proxy to evaluate if the particle can conduct all the energy it absorbed. In LII particle cooling monitoring, a similar observation has been associated with the Sauter mean diameter of the particle (Liu et al 2005b, Sipkens et al. 2014)– which is equivalent to the V/S ratio shown here and further affirms the validity of our model."

(7) As a supplement to Table 1, it may be worth having a table stating what properties are used in the model (e.g., thermal accommodation coefficient, density, specific heat, etc.). This will make clear which parameters were used in the model and how they compare to others in the literature.

Thank you for making this comment. We have added a few rows in Table 1 for the input parameters that you specified.

(8) Instead of Table 1, the authors should also consider adding temperature-time decays to show how the models compare. The authors could also add the diameter and heat transfer modes, in separate panels, if they wanted.

We appreciate this comment, but we prefer not to add separate panels for the diameter and heat transfer modes.

(9) Could the authors show sample temporal signals in general, with both the experimental and simulated results. For example, show experimental data alongside the heat conduction profile results from Fig. 3. How well do these signals match? This would be a far better test of the model and may show some of the reason for remaining discrepancies in Fig.4, which is currently unexplored.

Thank you for your comment. This is not possible for Figure 3 and 4 as we only have a temporal profile for the simulated data and not for the experimental. The experimental data available is only the sound intensity, aka the measured concentration, for each light modulation profile.

(10) Why did the authors choose to normalize by a 20 nm spherical particle in Sec. 3.3 (e.g., it is far enough into the free molecular conduction and Rayleigh absorption regimes). What if the authors chose a smaller size to ensure these two conditions are satisfied?

We have added two references that suggest this as a typical size for BC particles. Specifically, Bond, Light absorption by carbonaceous particles: An investigative review, 2006 and Olsen, Universal relations between soot effective density and primary particle size for common combustion sources, 2019.

(11) Could the authors clarify (i.e., remind the reader) how the fractal dimension is implemented into the simulations? To which fractal dimension are the authors referring?

We have clarified both points. We obviously refer to 3D fractal dimension. We have added the following:

“Particle morphology is examined by varying the three-dimensional fractal dimension between 1.2 and 2.8 extending beyond the typical range of 1.8 – 2.8 (Olfert and Rogak, 2019), to check the consistency of the results.”

(12) The signal decrease observed in Fig. 6 assumes a constant morphology, which will undoubtedly change between BC and ash. Further, similar trends may result in some of these features being correlated and thus difficult to distinguish from another. The authors should, at a minimum, note this caveat and weaken the statement.

It is noted that the authors add a caveat to the effect of the above statement in a later section, but the current phrasing and flow could be improved. Do all of these trends need to be shown when they are mathematically equivalent? Could the authors instead spend the time showing that they are mathematically equivalent (i.e., the chosen dimensionless time yields a compact equation that applies universally).

We restructured the information presented, bringing the caveat of V/S limitation caveat in section 3.4 to improve the flow, as suggested by the reviewer.

With regard to the suggestion by the reviewer on mathematical equivalency, we still believe that explaining the approach step by step and building on the different particle properties is a more educational approach that validates our final conclusion. We hope that this approach is still acceptable by the reviewer. The added section is:

“. For example, a particle with very high heat capacity will absorb more energy than it can conduct to the surrounding air, leading to reduced normalised signal. However, physical properties alone cannot be used to normalize the signal from devices of different operation characteristics or to correct for different V/S ratios. Thus, we propose a better metric in section 3.6.”

(13) Could the authors propose a different quantity than “TOA”, given the common use of the acronym in reference to thermal optical analysis in overlapping literature. Also consider using the symbol tau for a dimensionless time decay.

Thank you for highlighting this. We changed it to Tc which denotes the cooling time

(14) The paper could benefit from a practical implementation dimensionless quantity calculated for some of the proposed particle classes, to show how they might differ or be used to correct measurements.

We would like to thank the reviewer for this comment. Even though it would indeed be beneficial we decided not to proceed with this.

(15) A key limitation of the current paper is that the applied model is rather simplistic, despite trying to capture the complex effect of fractal morphology. For example, the authors do not consider polydispersity, which disrupts the simple collapse of the data that the authors observed. This may lead to overly optimistic results, which cannot be easily validated against experiments (which would require precise control of the fractal dimension for particles of the same size). The paper is then too confident in the correction that can be applied. The manuscript remains useful, as the brief comparison to experimental data shows, but the conclusions from the simulations must be made less confident. Appropriate caveats need to be added that this is a rather simple treatment (which is both an advantage and a limitation).

Thank you for your comment. You are right that the model is rather simple and we consider this an advantage for our needs which is the ability to run multiple simulations in literally seconds and understand the phenomena in play and the ways they can be exploited by sensing systems. Polydispersity is indeed an source of error as the phenomena involved are not linear. We have added the following statement at the end of the conclusions section:

“Finally, there are two main caveats associated with the method proposed. The approach proposed remains rather simple with regard to the absorption coefficient calculation, that could be improved using a more detailed model or specific BC absorption databases (Liu et al. 2019). In addition, aerosol polydispersity in real applications adds a degree of freedom (e.g. standard deviation of size distribution), potentially complicating signal interpretation."

(16) "… optoacoustic (OA) and optothermal (OT) ones and Laser Induced Incandescence (LII) …" Like the other techniques, "laser-induced incandescence" does not need to be capitalized.

Thank you for your comment. We changed to lowercase letters for all techniques.