This paper attempts to evaluate the impact of the uncertainty of black carbon refractive index on optical thickness and radiative forcing effects, which is a very valuable topic. However, the paper fails to thoroughly discuss the impact of the uncertainty of refractive index. Here are my comments:

We thank the reviewer for their very helpful feedback on our manuscript. Our responses to their comments are provided in green, below.

(1) This paper only tests three refractive indices, whereas in reality, the refractive index of black carbon is far more complex. Firstly, the refractive index of black carbon is spectrally dependent, which the authors have not elaborated on in detail. Secondly, the refractive index of black carbon may vary within a wider range. Even when adopting the maximum refractive index suggested by Bond et al. (2006), the mass absorption cross-section simulated by the current model falls below the lower limit of the observed range. Liu et al. (2020) suggested higher refractive index values by comparing the differences between simulations and measurements.

References:

Bond, T. C., & Bergstrom, R. W. (2006). Light Absorption by Carbonaceous Particles: An Investigative Review. Aerosol Science and Technology, 40 (1),27-67.

Liu, L., & Mishchenko, M. I. (2005). Effects of aggregation on scattering and radiative properties of soot aerosols. Journal of Geophysical Research: Atmospheres,495 110 (D11).

Liu, F., Yon, J., Fuentes, A., Lobo, P., Smallwood, G. J., & Corbin, J. C. (2020). Review of recent literature on the light absorption properties of black carbon: Refractive index, mass absorption cross section, and absorption function. Aerosol Science and Technology, 54 (1), 33-51.

Kahnert, M. (2010). On the discrepancy between modeled and measured mass absorption cross sections of light absorbing carbon aerosols. Aerosol Science and Technology, 44 (6), 453-460.

Luo, J., Zhang, Y., Wang, F., & Zhang, Q. (2018). Effects of brown coatings on the absorption enhancement of black carbon: a numerical investigation. Atmospheric Chemistry and Physics, 18 (23), 16897–16914.

Regarding the choice of BCRI assessed in this work: we agree with the reviewer that even the “high” Bond and Bergstrom (2006) scheme underestimates BC absorption. Our original manuscript restricted its main analysis to BCRI commonly used in Earth system models, and only touched on the impacts of using more strongly-absorbing schemes in the discussion section. In the revised manuscript, we have moved the Bescond et al. (2016) refractive index, which was recommended by Liu et al. (2020), from the discussion to our main analysis. In the process we have renamed our schemes from “low”, “medium”, and “high” to “dA1991”, “BB2006low”, “BB2006high”, and “Besc2016” for improved clarity. The Besc2016 results are included in all of our figures. In brief, increasing BC absorption from the dA1991 to BB2006low (BB2006high, Besc2016) BCRI scheme increases global-mean 2015-2019 AAOD by 27% (42%, 59%) and ERFari by 32% (47%, 100%).

We then address a number of other BCRI schemes in the discussion [lines 402-419]:

> *We have assessed four BCRI schemes here, but many others exist. As well as the Bescond et al. (2016) scheme assessed here, the review by Liu et al. (2020) highlighted the Williams et al. (2007) values of m_{635nm} = 1.75-1.03i and E(m_{635nm}) = 0.365 as being consistent with current estimates of the absorption function. Williams et al. (2007) reported measurements at 635nm and 1310nm and did not extrapolate to other wavelengths, but assuming a relatively flat E(m) through the visible range, this would indicate a degree of absorption somewhere between our BB2006high and Besc2016 schemes. Other estimates which have been widely used in the combustion science literature, such as the Janzen (1979) value of m=2.0-1.0i for all visible wavelengths or the more recent Moteki et al. (2010) m_{1064nm} = (2.26±0.13) – (1.26±0.13)i, also yield E(m) between BB2006high and Besc2016, but with values low enough that they are not recommended by Liu et al. (2020).*

Refractive indices determined from laboratory measurements may not be representative of atmospheric black carbon. For instance, BC generated by a simple, clean laboratory flame will likely have a different temperature history – and thus, different optical and structural properties – than that generated by the more complex sources responsible for most atmospheric BC (Bond and Bergstrom, 2006). Combustion experiments also measure freshly emitted particles, which may have substantial morphological differences from hours-to-days old atmospheric BC. The recent work by Moteki et al. (2023), who measured the refractive indices of atmospheric BC particles sampled during a scientific cruise in the northwest Pacific may be more suitable for use in climate models. By combining their optical measurements with the constraints imposed by the accepted mass absorption cross section of BC, they obtained a range of plausible refractive indices suitable for describing atmospheric BC. Their recommended value, $m_{633nm} = 1.95 - 0.96i$ with $E(m_{633nm}) = 0.297$, also falls between the BB2006high and Besc2016 schemes.

Regarding the spectral dependence of the refractive index: we have expanded our description of the spectral characteristics of the four BCRI schemes [lines 141-144]:

At all wavelengths, the dA1991 scheme has the lowest absorption and the Besc2016 the highest. E(m) in the first three schemes increase to both the ultraviolet and infrared, while the Besc2016 scheme decreases slightly to the infrared, but all four are fairly constant through the visible and near-infrared (d'Almeida et al., 1991; Chang and Charalampopoulos, 1990; Bescond et al., 2016).

We have also clarified in the abstract that we vary the full BCRI spectrum, not only its value at 550nm [line 5]:

*With other parameterizations held constant, changing BC's **spectrally-varying** refractive index from the least- to most-absorbing estimate commonly used in Earth system models…*

Our analysis is focused on 550nm, because that is the wavelength for which Earth system models typically publish aerosol optical data. However, we have included references to a now-published companion manuscript, Li et al. (2024), which examines three of our four BCRI (dA1991, BB2006high, and Besc2016) in an offline radiative transfer model. Their work includes assessment of the impacts of BCRI on the spectral dependence of BC optical properties. We clarify this in lines 146-151:

Although our experiments vary the BCRI at all wavelengths, our analysis predominantly focuses on the characteristics of these schemes at 550nm. This is the wavelength for which Earth system models typically publish aerosol optical data and for which many satellite retrievals are available. In a complementary analysis, Li et al. (2024) assess the optical properties of BC in the dA1991, BB2006high, and Besc2016 schemes, including dependence on wavelength, particle size, and mixing state. The Li et al. (2024) analysis relies on theoretical calculations and a one-dimensional radiative transfer model, while the work presented here explores the impacts of the BCRI on Earth system model simulations.

References:

Bescond, A., Yon, J., Ouf, F.-X., Rozé, C., Coppalle, A., Parent, P., Ferry, D., and Laffon, C.: Soot optical properties determined by analyzing extinction spectra in the visible near-UV: Toward an optical speciation according to constituents and structure, Journal of Aerosol Science, 101, 118–132, https://doi.org/10.1016/j.jaerosci.2016.08.001, 2016.

Bond, T. C. and Bergstrom, R. W.: Light Absorption by Carbonaceous Particles: An Investigative Review, Aerosol Science and Technology, 40, https://doi.org/10.1080/02786820500421521, 2006.

Janzen, J.: The refractive index of colloidal carbon, Journal of Colloid and Interface Science, 69, 436–447, https://doi.org/10.1016/0021- 9797(79)90133-4, 1979.

Li, J., Digby, R., and von Salzen, K.: Accounting for black carbon refractive index in atmospheric radiation, Quarterly Journal of the Royal Meteorological Society, pp. 1–14, https://doi.org/10.1002/qj.4842, 2024.

Liu, F., Yon, J., Fuentes, A., Lobo, P., Smallwood, G. J., and Corbin, J. C.: Review of Recent Literature on the Light Absorption Properties of Black Carbon: Refractive Index, Mass Absorption Cross Section, and Absorption Function, Aerosol Science and Technology, 54, https://doi.org/10.1080/02786826.2019.1676878, 2020.

Moteki, N., Kondo, Y., and Nakamura, S.-i.: Method to measure refractive indices of small nonspherical particles: Application to black carbon particles, Journal of Aerosol Science, 41, 513–521, https://doi.org/https://doi.org/10.1016/j.jaerosci.2010.02.013, 2010.

Moteki, N., Ohata, S., Yoshida, A., and Adachi, K.: Constraining the complex refractive index of black carbon particles using the complex forward-scattering amplitude, Aerosol Science and Technology, 57, 678–699, https://doi.org/10.1080/02786826.2023.2202243, 2023.

Williams, T., Shaddix, C., Jensen, K., and Suo-Anttila, J.: Measurement of the dimensionless extinction coefficient of soot within laminar diffusion flames, International Journal of Heat and Mass Transfer, 50, 1616–1630, https://doi.org/10.1016/j.ijheatmasstransfer.2006.08.024, 2007.

(2) The author's selection and simulation of refractive indices have not been rigorously discussed in conjunction with measurements.

We have expanded our descriptions of the four BCRI schemes. They now include additional description of the historical and measurement context (for example, the fact that dA1991 was included in the OPAC tables and entered widespread use in the climate modelling community, or the fact that BB2006high is known to be insufficiently absorbing and that this cannot be explained by the choice of optical model alone). This context should help to clarify our motivation for selecting the schemes we did. Each scheme description also now includes explicit mention of the assumed optical model and BC density. [Lines 85-135]

(3) The morphology of black carbon and the choice of model can affect the uncertainty introduced by the refractive index, yet the author has not elaborated on the model used in the simulations, which I presume to be spherical. Additionally, the mixing state and particle size distribution also contribute to the uncertainty stemming from the refractive index, yet the author has not comprehensively discussed these factors in conjunction with measurements. Instead, default values from the model were utilized.

References:

Luo, J., Li, D., Wang, Y., Sun, D., Hou, W., Ren, J., . . . Qiu, J. (2023). Quantifying the effects of the microphysical properties of black carbon on the determination of brown carbon using measurements at multiple wavelengths, Atmos. Chem. Phys., 24, 427–448, https://doi.org/10.5194/acp-24-427-2024, 2024.

Luo, J., Li, Z., Zhang, C., Zhang, Q., Zhang, Y., Zhang, Y., . . . Chakrabarty, R. K. (2022). Regional impacts of black carbon morphologies on short wave aerosol–radiation interactions: a comparative study between the US and China. Atmospheric Chemistry and Physics, 22 (11), 7647–7666.

Luo, J., Zhang, Q., Luo, J., Liu, J., Huo, Y., & Zhang, Y. (2019). Optical modeling of black carbon with different coating materials: The effect of coating configurations. Journal of Geophysical Research: Atmospheres, 124 (23), 13230-13253.

We thank the reviewer for raising these concerns. In recognition of its importance for interpreting the manuscript, we have both expanded our description of BC optics in CanAM5.1-PAM and moved the model description from the appendix to the main text. In particular, we have expanded the description of aerosol optical calculations to read [lines 181-191],

> *Aerosol optical properties in PAM are determined from pre-computed lookup tables. The tables are generated using Mie theory to determine optical properties as a function of relative humidity, wavelength, and particle size. Although the assumption of spherical particles may be problematic for freshly-emitted BC, the majority of the BC simulated by an Earth system model is hours to days old and will be relatively compact and/or internally mixed. Mie theory is thus a reasonable approximation. For internally mixed aerosol an effective refractive index is computed using the Maxwell-Garnett*

approximation (Wu et al., 2018), which has been demonstrated to describe the optical properties of coated BC better than volume-weighted or core-shell approximations (Adachi et al., 2010; Stevens and Dastoor, 2019).

Other treatments of aerosol morphology and mixing state would likely yield different absolute values of simulated AAOD and ERFari. However, the focus of this work is on the difference in these quantities between simulations conducted with different BCRI schemes, with other parameterizations held constant. As it is, CanAM5.1-PAM's simulated AAOD is in good agreement with other Earth system models and with observations…

We focus on the BCRI only, without also investigating the impacts of uncertainty in BC morphology or mixing state, precisely because of the challenge in disentangling the different drivers of simulated absorption across different models using different treatments of these calculations, for example as seen in Sand et al. (2021).

References:

Adachi, K., Chung, S. H., and Buseck, P. R.: Shapes of soot aerosol particles and implications for their effects on climate, Journal of Geophysical Research: Atmospheres, 115, https://doi.org/10.1029/2009JD012868, 2010.

Sand, M., Samset, B. H., Myhre, G., Gliß, J., Bauer, S. E., Bian, H., Chin, M., Checa-Garcia, R., Ginoux, P., Kipling, Z., Kirkevåg, A., Kokkola, H., Le Sager, P., Lund, M. T., Matsui, H., van Noije, T., Olivié, D. J. L., Remy, S., Schulz, M., Stier, P., Stjern, C. W., Takemura, T., Tsigaridis, K., Tsyro, S. G., and Watson-Parris, D.: Aerosol Absorption in Global Models from AeroCom Phase III, Atmospheric Chemistry and Physics, 21, https://doi.org/10.5194/acp-21-15929-2021, 2021.

Stevens, R. and Dastoor, A.: A review of the representation of aerosol mixing state in atmospheric models, Atmosphere, 10, https://doi.org/10.3390/atmos10040168, 2019.

Wu, K., Li, J., von Salzen, K., and Zhang, F.: Explicit solutions to the mixing rules with three-component inclusions, Journal of Quantitative Spectroscopy and Radiative Transfer, 207, 78–82, https://doi.org/10.1016/j.jqsrt.2017.12.020, 2018.

(4) The author's introduction to the optical simulation of black carbon in the model is overly simplistic, failing to comprehensively study the impact of black carbon uncertainty on optical properties and discuss it in conjunction with measurements.

As described for comment 3, above, we have expanded our description of the optical simulation of BC in the model [lines 179-185]. Although the question of how uncertainties in BC morphology and mixing state impacts its simulated optical properties is an interesting one, it is outside the scope of this analysis. Our focus is on the impact that the choice of refractive index has on simulated climate-relevant fields such as AAOD and ERFari.

To address the uncertainty of BC optical properties in conjunction with measurements, we have expanded our description of sources of uncertainty in laboratory measurements of the BCRI, including the inversion of measurements to obtain spectrally varying information [lines 54-58]:

The measurements are then inverted to yield the full spectrally-varying refractive index, for example using the Kramers-Kronig relations (Chang and Charalampopoulos, 1990) or the Drude-Lorentz dispersion relation (Dalzell and Sarofim, 1969; Lee and Tien, 1981). The former method is exact, but requires measurements over a greater range of wavelengths; the latter requires fewer measurements but yields poor results at visible wavelengths (Chang and Charalampopoulos, 1990; Bond and Bergstrom, 2006; Menna and D'Alessio, 1982).

And more description of the uncertainty stemming from the choice of optical model [lines 59-74]:

The choice of optical model can introduce substantial uncertainty into the derived refractive index. Historically, many estimates of the refractive index of black carbon have used Mie theory (Mie, 1908) which provides an exact solution to Maxwell's equations for scattering from spherical particles. However, freshly emitted BC particles are not spherical, but instead consist of fractal-like aggregates of individual monomers. Assuming Mie theory can result in substantial *underprediction of these particles' absorption and scattering; furthermore, the inferred BCRI describes a combination of pure BC and the air contained within the aggregates' voids, whereas the objective is to measure the refractive index of pure BC (Bond and Bergstrom, 2006). Improving on Mie theory, a number of optical models for aggregate particles exist. Rayleigh-Debye-Gans (RDG) theory remains the most frequently used due to its simplicity (Liu et al., 2020), but it provides an approximate solution only and does not account for multiple scattering between the monomers, which may lead to underestimation of the mass absorption cross section (Mackowski, 1995; Bond and Bergstrom, 2006). Numerically exact solutions for aggregate particles include the multi-sphere T-matrix (MSTM; Mackowski and Mishchenko, 1996) and generalized multi-particle Mie (GMM; Xu, 1995) methods for aggregates composed of non-overlapping spheres, or the discrete dipole approximation (DDA; Purcell and Pennypacker, 1973; Yurkin and Hoekstra, 2007) for more complex morphologies. For more on these methods, the interested reader is referred to Kahnert and Kanngießer (2020). In practice, however, the RDG approximation is often sufficient for BC since its scattering is so low and other uncertainties are so high (Kahnert and Kanngießer, 2020; Mackowski, 1995). The four BCRI schemes assessed in this work were derived using Mie theory or RDG-type optical models.*

References:

Bond, T. C. and Bergstrom, R. W.: Light Absorption by Carbonaceous Particles: An Investigative Review, Aerosol Science and Technology, 40, https://doi.org/10.1080/02786820500421521, 2006.

Chang, H.-C. and Charalampopoulos, T. T.: Determination of the wavelength dependence of refractive indices of flame soot, Proceedings of the Royal Society of London. Series A: Mathematical and Physical Sciences, 430, 577–591, https://doi.org/10.1098/rspa.1990.0107, 1990.

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Liu, F., Yon, J., Fuentes, A., Lobo, P., Smallwood, G. J., and Corbin, J. C.: Review of Recent Literature on the Light Absorption Properties of Black Carbon: Refractive Index, Mass Absorption Cross Section, and Absorption Function, Aerosol Science and Technology, 54, https://doi.org/10.1080/02786826.2019.1676878, 2020.

Mackowski, D. W.: Electrostatics analysis of radiative absorption by sphere clusters in the Rayleigh limit: application to soot particles, Applied Optics, 34, 3535–3545, https://doi.org/10.1364/AO.34.003535, 1995.

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Mie, G.: Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen, Annalen der Physik, 330, 377–445, https://doi.org/https://doi.org/10.1002/andp.19083300302, 1908.

Purcell, E. M. and Pennypacker, C. R.: Scattering and Absorption of Light by Nonspherical Dielectric Grains, Astrophysical Journal, 186, 705–714, https://doi.org/10.1086/152538, 1973.

Xu, Y.: Electromagnetic scattering by an aggregate of spheres, Applied Optics, 34, 4573–4588, https://doi.org/10.1364/AO.34.004573, 1995.

Yurkin, M. and Hoekstra, A.: The discrete dipole approximation: An overview and recent developments, Journal of Quantitative Spectroscopy and Radiative Transfer, 106, 558–589, https://doi.org/10.1016/j.jqsrt.2007.01.034, iX Conference on Electromagnetic and Light Scattering by Non-Spherical Particles, 2007.

In summary, the current version of this manuscript is overly simplistic, testing only a few refractive index cases without adequately discussing the uncertainty in refractive index, the discrepancies between simulations and measurements, how to interpret measurements through refractive index adjustments, the spectral dependence of refractive index, the impact of microphysical properties on refractive index uncertainty, and its consequences on radiative forcing.

We thank the reviewer for identifying these gaps in our discussion of the material. It is our sincere hope that with our expanded text and the addition of the Bescond et al. (2016) refractive index to our main analysis, this manuscript will now be to their satisfaction.