

General comments

This manuscript proposes a revised framework for classifying organic aerosols using wavelength-dependent absorption properties. The authors compile literature values of complex refractive indices and related physical parameters for selected OA species, classify OA into very weakly, weakly, moderately, and strongly absorbing categories, and calculate corresponding MAC and SSA values using Mie theory. The broader motivation is to improve the representation of OA optical properties in climate models, satellite retrievals, and radiative-transfer calculations.

However, the current work is mainly a literature-based optical-parameter compilation and classification exercise. The proposed framework has some conceptual value, but it does not yet provide a clear pathway for use in real atmospheric studies or models. In particular, the manuscript does not explain how users should determine the abundance or mass fraction of each proposed OA class for a given time, region, source mixture, altitude, or aging state. This missing step severely limits the practical value of the proposed classification for radiative forcing, satellite retrieval, or climate-model applications.

I therefore believe that the manuscript would require a fundamental revision and substantial redevelopment before it could be considered for publication. If an operational abundance-assignment method and application test cannot be provided, the work may be framed explicitly as a conceptual, perspective-like, or technical/database contribution rather than as an already deployable research framework.

Specific comments

1. The most important limitation is that the manuscript provides optical properties for selected surrogate species, but not a practical method for assigning ambient OA to the proposed classes. For radiative-effect calculations, modelers and observational researchers need to know not only the RI, MAC, and SSA of S-BrC, M-BrC, W-BrC, and VW-BrC, but also the amount of OA in each class for the specific atmosphere being studied. For example, users would need to know the relative abundance of these classes in wintertime urban China, biomass-burning plumes over the Amazon, aged wildfire smoke over the free troposphere, biogenic SOA-dominated forest regions, or polluted continental outflow. The manuscript does not provide such spatiotemporally resolved class abundances, nor does it explain how they can be obtained. This is a critical gap. Without a mapping from real atmospheric OA to the proposed optical classes, the framework cannot be directly used for regional or global radiative forcing calculations. At present, the manuscript appears to provide an optical lookup table rather than a complete classification framework that can be implemented in models or applied to observations.

Response 1: *We thank the reviewer for pointing this out. The reviewer raises a valid and important concern regarding the applicability of the aerosol classification presented in this study to climate and weather models.*

The primary aim of this work is to address a critical gap in the representation of organic aerosols. Most current studies and climate models treat organic aerosols as predominantly

scattering species, although recent improvements have enabled the representation of absorbing organic aerosols in some models. Our study highlights the importance of representing organic aerosols as mixtures originating from multiple sources, rather than treating them as a single homogeneous species, particularly for aerosol types such as biomass-burning smoke and urban pollution.

We agree with the reviewer that understanding the relative abundance of different organic aerosol components and establishing a framework for defining aerosol types are essential for their improved representation in climate and weather models. We intend to address this aspect in our subsequent work. In that study, we will compile organic aerosol mass fractions reported from multiple field campaigns and use these measurements to determine the optical properties of different smoke and urban aerosol mixtures. This approach will provide a more physically representative framework for deriving optical parameters for aerosol types that can ultimately be incorporated into atmospheric and climate models.

Thus, while the present study focuses on establishing the importance of accounting for the optical diversity of organic aerosols, our subsequent work will build upon these findings by incorporating observationally constrained organic aerosol mass fractions to develop model-relevant optical properties for different aerosol types.

We have added the following text to the “Caveats and Limitations” section of the manuscript.

“The primary aim of this work is to address a critical gap in the representation of organic aerosols. Most current studies and climate models treat organic aerosols as predominantly scattering species, although recent improvements have enabled the representation of absorbing organic aerosols in some models. Our study highlights the importance of representing organic aerosols as mixtures originating from multiple sources, rather than treating them as a single homogeneous species, particularly for aerosol types such as biomass-burning smoke and urban pollution.

Although, for the framework to be used in global and regional radiative transfer model users need to compile organic aerosol mass fractions reported from multiple field campaigns and use these measurements to determine the optical properties of different smoke and urban aerosol mixtures. This approach will provide a more physically representative framework for deriving optical parameters for aerosol types that can ultimately be incorporated into atmospheric and climate models.

Thus, while the present study focuses on establishing the importance of accounting for the optical diversity of organic aerosols, subsequent work will build upon these findings by incorporating observationally constrained organic aerosol mass fractions to develop model-relevant optical properties for different aerosol types.”

2. The manuscript currently gives the impression that the proposed classification and TAO database can serve as a broadly applicable tool for climate models, satellite retrievals, and field campaigns. This claim is too strong in its current form. The work may be useful as a preliminary optical-parameter compilation or as a conceptual framework for organizing OA absorption strength. However, it is not yet a complete database for atmospheric radiative applications.

I recommend that the authors substantially revise the framing. The manuscript should explicitly state that the proposed classes are optical endmembers or representative surrogate classes, not a complete description of real-world OA spatiotemporal distributions. The authors should avoid implying that the framework can already be directly applied to global radiative forcing estimates unless they demonstrate such an application.

Response: *We thank the reviewer for this insightful comment. Organic aerosols are commonly represented in climate models using a single particle refractive index or a simplified set of optical properties. Our study demonstrates that organic aerosol species can exhibit a broad range of optical behavior, from highly scattering to strongly absorbing. Therefore, representing organic aerosols using a single optical property may introduce substantial biases in model outputs.*

We agree with the reviewer that the characterization of aerosol optical properties by aerosol type, rather than solely by individual species, is particularly important for satellite retrievals and field campaigns, where the optical properties of complex mixtures such as biomass-burning smoke and urban pollution need to be accurately represented. Thus, reporting the optical parameters of different aerosol types, in addition to those of individual species, is essential.

We are addressing this broader need through our TAO database, which will provide optical parameters for both individual aerosol species and different aerosol types. These data can be directly utilized in climate modeling, satellite retrievals, and field-campaign studies. To clarify the intended applications of the present study and avoid potential confusion, we have therefore rephrased the relevant text as follows:

Old version: “This choice of species includes both absorbing and scattering OA and enables more accurate representation of OA in climate models and satellite retrievals, improving aerosol radiative forcing estimates.”

New version: “This choice of species that includes both absorbing and scattering OA along with their mass fraction and size distribution measurements from the field will allow more accurate modeling of biomass burning smoke and urban optical parameter computations in climate models and satellite retrievals, improving aerosol radiative forcing estimates.”

3. The classification into VW-BrC, W-BrC, M-BrC, and S-BrC appears to be closely related to previous absorption-strength frameworks, particularly those based on volatility and imaginary refractive index. The manuscript extends this idea across multiple wavelengths and compiles selected surrogate values, but the conceptual novelty is not sufficiently clear.

The authors should more explicitly state what is new relative to previous work. Is the novelty the wavelength-dependent thresholds? The selection of representative surrogates? The conversion from IRI to MAC and SSA? The intended inclusion in TAO? If so, the authors need to demonstrate why these additions constitute a substantial advance rather than an incremental reorganization of existing literature values.

At present, the work lacks a clear independent validation or application showing that the proposed scheme improves model performance, retrieval interpretation, or radiative forcing estimates relative to existing OA representations such as OPAC or previous BrC parameterizations.

Response: *As noted by the reviewer, the terminology used for the classification was adapted from previous studies; however, the classification approach presented in our study is novel in several important aspects.*

- 1. Wavelength-dependent classification: Our study demonstrates that the classification of organic aerosols strongly depends on the wavelength at which their optical properties are evaluated. We therefore provide a wavelength-dependent classification and recommend optimal wavelengths for distinguishing organic aerosols into the four proposed categories. We also provide wavelength-dependent thresholds for defining each class, which can be applied to measurements or model simulations for calculating radiative effects.*
- 2. Integration of physical, chemical, and optical properties: While we demonstrate that IRI is a critical parameter for separating organic species classes, we also provide a framework for assigning mass absorption coefficients (MACs) and single scattering albedo (SSA) values for the same organic classes. This novel contribution provides a direct connection between the proposed optical categories and the organic aerosol species that are present in the atmosphere for model simulations and measurement closure studies. These MAC and SSA values are also wavelength dependent. To better highlight these contributions, we have added the following sentence to the manuscript:*

We have added the following text into the conclusion section of the manuscript:

“This study builds on previous work to provide another step towards improving measurement-model synergy. Our approach expands current schemes for classifying organic aerosols by considering physical, chemical, and optical properties together. Most importantly, this work provides:

Wavelength-dependent classification: Our study demonstrates that the classification of organic aerosols strongly depends on the wavelength at which their optical properties are evaluated. We therefore provide a wavelength-dependent classification and recommend optimal wavelengths for distinguishing organic aerosols into the four proposed categories. We also provide wavelength-dependent thresholds for defining each class, which can be applied to measurements or model simulations for calculating radiative effects.

Integration of physical, chemical, and optical properties: While we demonstrate that IRI is a critical parameter for separating organic species classes, we also provide a framework for assigning mass absorption coefficients (MACs) and single scattering albedo (SSA) values for the same organic classes. This novel contribution provides a direct connection between the proposed optical categories and the organic aerosol species that are present in the atmosphere for model simulations and measurement closure studies. These MAC and SSA values are also wavelength dependent.”

4. The manuscript first classifies OA into BrC and scattering OA, but then assigns biogenic SOA and anthropogenic SOA to VW-BrC and W-BrC categories. This creates conceptual confusion. If a species is essentially scattering, has SSA close to 1, has very low MAC, and has an imaginary refractive index near or below detection limits at visible wavelengths, it is not clear that it should be called brown carbon in the usual atmospheric sense. The use of “VW-BrC” may be defensible as an operational optical class, but the manuscript needs to make this distinction explicit.

I recommend that the authors consider alternative terminology such as “very weakly absorbing OA” and “weakly absorbing OA,” rather than labeling all low-absorption OA as BrC. If the authors retain the BrC terminology, they should clearly explain that VW-BrC and W-BrC are operational absorption-strength classes and do not necessarily imply that all included species would be identified as brown carbon in the traditional chemical or observational sense.

Response: *We thank the reviewer for noting this important point. We agree that the term “BrC” should be used carefully when the magnitude of light absorption is very small. However, previous studies have commonly defined BrC based on the presence of wavelength-dependent light absorption in the UV–visible region, rather than solely on the magnitude of absorption. In our study, all species classified as BrC exhibit a clear wavelength dependence in their absorption spectra, although the absorption strength varies substantially among species. Therefore, we retain the term BrC and use the VW-BrC and W-BrC categories to distinguish species according to the strength of their wavelength-dependent absorption.*

We have added the following text in the manuscript to clarify this point:

“Note that our classification nomenclature is based on a continuum of absorption strength, from very-weakly (VW) to strongly (S), for organic aerosols. Since we show that this absorption is always wavelength dependent, we use the term brown carbon “BrC” to represent the different classes.”

5. The manuscript selects a small number of surrogate species for each class, including fresh and aged tarballs, acetone-extracted OA, SRFA, toluene-derived SOA, and limonene or α -pinene-derived SOA. These choices are partly reasonable, but the justification is not sufficient for a framework intended to serve broad atmospheric applications.

The authors should provide a more systematic rationale for surrogate selection and discuss the representativeness and limitations of each surrogate.

Response: As noted by the reviewer, we have added 2 columns to table 6 to make the representativeness and limitations clearer.

Table 6. RI, MAC, and SSA for the species selected to represent each class in TAO.

Class	Wavelength	Refractive Index	MAC	SSA	Representativeness	Limitation
Fresh Tarballs	300	$1.32 + 0.13i$	1.44	0.54	Highly absorbing wiBrC	The RI is derived after passing a high-energy electron beam, causing volatile substances to evaporate. Hence, while representing highly absorbing BrC, tarballs should be mixed with wsBrC.
	350	$1.33 + 0.11i$	1.38	0.56		
	400	$1.33 + 0.093i$	1.26	0.60		
	450	$1.33 + 0.082i$	1.15	0.63		
	500	$1.32 + 0.072i$	1.02	0.66		
	550	$1.32 + 0.060i$	0.87	0.70		
Aged Tarballs	300	$1.35 + 0.11i$	1.42	0.55	Aged absorbing wiBrC	The surrogate may not fully capture the chemical evolution of tarballs during atmospheric aging and may not represent the full range of aged tarball properties.
	350	$1.37 + 0.088i$	1.32	0.60		
	400	$1.37 + 0.073i$	1.18	0.65		
	450	$1.37 + 0.057i$	0.99	0.71		
	500	$1.38 + 0.045i$	0.82	0.76		
	550	$1.38 + 0.033i$	0.62	0.82		
wiBrC (Acetone extracted)	350	$1.67 + 0.17i$	4.65	0.60	Strongly absorbing wiBrC	Acetone extraction may alter the composition of the original BrC and may not represent the complete atmospheric population of wiBrC.
	400	$1.67 + 0.11i$	3.53	0.68		
	450	$1.67 + 0.063i$	2.18	0.79		
	500	$1.67 + 0.045i$	1.50	0.84		
	550	$1.67 + 0.030i$	0.94	0.88		
wsBrC (SRFA)	400	$1.68 + 0.057i$	2.29	0.80	A mixture of wsBrC and wiBrC can represent atmospheric OA emitted from biomass burning	SRFA is a specific humic-like material and may not capture the full chemical and optical diversity of atmospheric wsBrC.
	450	$1.65 + 0.0048i$	0.22	0.98		
	500	$1.65 + 0.0016i$	0.065	0.99		
Anthropogenic OA (Toluene)	300	$1.57 + 0.020i$	1.02	0.90	Anthropogenic OA representative of urban pollution	Toluene-derived aerosol represents only one class of anthropogenic OA and may not capture the diversity of anthropogenic emissions and atmospheric processing.
	350	$1.56 + 0.0062i$	0.34	0.96		
	400	$1.55 + 0.0020i$	0.097	0.99		
	450	$1.54 + 0.0008i$	0.033	0.99		
	500	$1.53 + 0.0006i$	0.010	0.99		
	550	$1.53 + 0.0003i$	0.009	0.99		
Biogenic OA (Limonene)	300	$1.55 + 0.0009i$	0.062	0.99	Biogenic OA with very weak light absorption to represent biogenic emissions from forests	Limonene-derived aerosol represents only one biogenic precursor and may not capture the diversity of biogenic OA from different precursors and oxidation pathways.
	350	$1.53 + 0.0003i$	0.012	0.99		
	400	$1.52 + 0.0002i$	0.008	0.99		
	450	$1.51 + 0.0001i$	0.004	0.99		
	500	$1.51 + 0.0001i$	0.003	0.99		
	550	$1.50 + 0.0002i$	0.005	0.99		

6. To establish practical significance, the authors should include at least one demonstration showing that the proposed framework improves something relative to existing OA

representations. Possible demonstrations may include: a radiative-transfer sensitivity calculation comparing OPAC-style OA, single non-absorbing OA, and the proposed multi-class OA representation; a regional or global model sensitivity test showing the impact of plausible class fractions on direct radiative forcing; comparison with independent field measurements of multi-wavelength MAC, SSA, or AAE; application to biomass-burning, urban, and biogenic-dominated cases to show how the classification performs under different source regimes; a satellite retrieval sensitivity test showing how the different OA classes affect inferred aerosol optical properties.

Without such a demonstration, the manuscript remains largely conceptual and does not provide enough evidence that the framework has substantial practical value.

Response: *We thank the reviewers for this comment. As the reviewer suggested, we computed direct radiative forcing using Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) code for OPAC-style OA, single non-absorbing OA, and a mixture of organics that represent smoke. The following section will be added to the manuscript.*

“To demonstrate the importance of OA classification in determining its radiative effect, we provide a case study using the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (ricchiazzi et al. 1998} to calculate the net radiative flux at the top of the atmosphere (TOA) (Figure 4). SBDART requires aerosol optical properties, including the single-scattering albedo (SSA), asymmetry parameter, and extinction efficiency, together with atmospheric and surface parameters such as solar zenith angle, season, and surface albedo. The difference in TOA net radiative fluxes between aerosol-loaded and aerosol-free conditions was used to calculate the TOA direct radiative forcing (TAO RF) associated with the aerosol layer.

TOA RF was calculated for four aerosol cases: (1) OPAC water-soluble OA, (2) OPAC water-insoluble OA, (3) biogenic OA represented as a non-absorbing aerosol, and (4) a mixture of organic and carbonaceous aerosol components representative of smoke. The smoke mixture was constructed based on the mass fractions of individual OA components reported by (posfai et al. 2003) for smoldering young smoke from Kruger National Park during the SAFARI 2000 field campaign. The mixture consisted of relative number concentrations of 17% black carbon (BC), 17% tarballs, and 66% scattering OA.

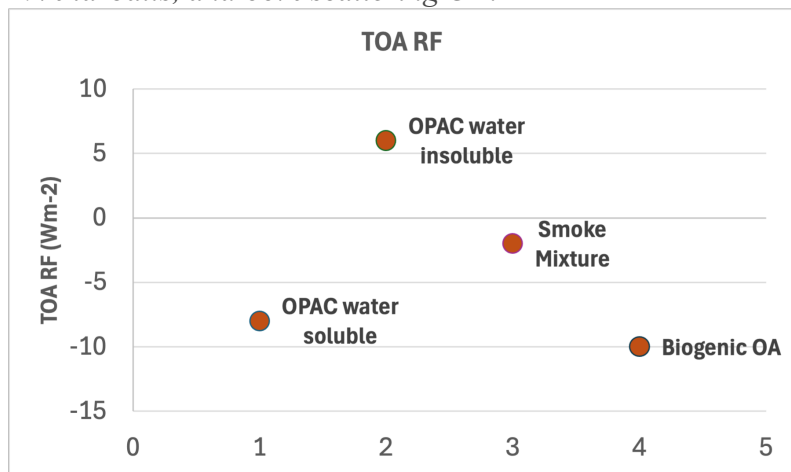


Figure: Top of the atmosphere radiative forcing calculations for a layer of aerosols in the atmosphere. The plot shows the radiative forcing for a single layer of: 1) OPAC water soluble, 2) OPAC water-insoluble, 3) Biogenic OA, and 4) Smoke Mixture.

The calculated TOA RF was -8 W m^{-2} for the OPAC water-soluble aerosol, $+6 \text{ W m}^{-2}$ for the OPAC water-insoluble aerosol, and -10 W m^{-2} for the non-absorbing biogenic OA. In contrast, the mixed smoke aerosol produced a TOA RF of -2 W m^{-2} . These results demonstrate that the assumed chemical composition and corresponding optical properties of OA can substantially influence the estimated radiative effect. Representing smoke using a single OA component may substantially overestimate or underestimate its net radiative forcing, depending on the assumed optical properties. Explicitly accounting for the contributions of scattering OA, tarballs, and BC therefore provides a more realistic representation of the radiative effects of smoke aerosol in climate models and radiative forcing calculations.”

Technical corrections

1. The manuscript should consistently distinguish OA from OC. The current discussion sometimes uses OA classification while Table 2 refers to “OC classification,” which may cause confusion.

Response: *We thank the reviewer for noting this discrepancy. We have corrected “OC classification” to “OA classification” in Table 2.*

2. The term “strongly absorbing BrC” should be carefully distinguished from black carbon. The manuscript should make clear that high BrC absorption does not imply BC-like refractoriness, morphology, lifetime, or source behavior.

Response: *In Figure 2, we have marked the IRI of BC with a black dotted line to distinguish it from the BrC absorption.*

3. Table 5: the W-BrC SSA range at 300 nm is written as “0.87-92”; this presumably should be “0.87-0.92”.

Response: *We thank the reviewer for catching this typo. We have corrected the SSA range as “0.87-0.92” in table 5.*

4. Throughout: define abbreviations at first use and use them consistently, including IRI, RI, MAC, SSA, TB, HULIS, SRFA, TAO, LVOC, SVOC, ELVOC, EELS, and AERONET.

Response: *We thank the reviewer for noting this. We have corrected all the double abbreviations.*

5. References: correct duplicated DOI prefixes such as “https://doi.org/https://doi.org/...”, standardize journal-title capitalization, complete incomplete bibliographic entries, and check typographical problems such as “De-scription”, “sub-stances”, “Berdowslci”, “Zandyelt”, and inconsistent capitalization of journal names.

Response: *We thank the reviewer for noting these typos. We have fixed them.*