This paper presents climatologies of aerosol absorption for 15 Aerocom models. The authors discuss the spread of the modeled results, and they compare modeled AAOD and AAE to AERONET. They find that modeled AAOD is biased low of AERONET, which has been a consistent finding for the lifetime of AERONET (e.g., *Sato et al.*, 2003; *Koch et al.*, 2009; *Bond et al.*, 2013). The paper is a light read and will probably be cited by the modeling community, but it does not provide the reader with new insight about why the model diversity exists, or why model AAODs continue to be biased low of AERONET AAODs (even 18 years after this was first noticed by *Sato et al.*, 2003).

The overarching goal of the paper is presented by the authors on Line 105:

"We aim to better quantify the sources of model spread by separating absorption per species (BC, OA, and dust) and investigate regional and seasonal differences."

Unfortunately, I do not believe that they accomplished this. They do separate absorption by species, but they do not use the same technique to compute species-specific absorption for the various models, and seemingly leave it up to the various modelers to submit their favorite technique for this comparison. This makes it very difficult for the reader to sort out the causes of the model spread that are presented in the paper, and indeed, the authors themselves seem to give up, attributing the model spread in MAC and AAOD on line 369 to *"…the complexities in separating the species and mixing assumptions where internal mixtures are assumed depending on how BC AAOD is calculated"*.

The authors recognize that computing species-specific absorption is easy for external mixtures, but at least nine of the models in Table 2 include internal mixing. Table 2 provides 1-2 sentences about how the models with internal mixing partition absorption amongst the individual species, but the descriptions are too brief for the reader to clearly understand what is being done. The main text does not help much, either. In the summary, the authors conclude on line 398 with: *"We recommend that the role of size and mixing rules and diagnostic procedures should be investigated in more detail to understand the differences in mass absorption coefficients."*. This leaves the reader wondering, though – why didn't the authors do that themselves already in this paper?

It would not be that difficult of a task to understand the spread in the MACs. The mass absorption coefficient (MAC) at any given wavelength for a pure species (i.e., external mix) is a function of the complex refractive index (mostly the imaginary component), size distribution, and density. For internally mixed aerosols, the volume fraction of the absorbing inclusions also plays a role. Thus, only 3 parameters are needed to analyze the MAC of external mixtures, and 4 paramaters are needed for internal mixtures. A 5th parameter that would be useful for analyzing the spread in species-specific column AAOD is column mass loading, but this is never mentioned in the article. The paper has the potential to make a significant contribution if the authors broke down the analysis into these 4-5 components. It would also be helpful to analyze the external and internal mixtures separately.

How to attribute absorption to different species for internal mixtures

The authors state that it is conceptually difficult to report separate absorption by species for internal mixtures (lines 85, 204, and 267). The conceptual difficulty occurs because the sum of the absorptions of the component species in an internal mixture does not equal the absorption of an internal mixture. The simplest example is a black carbon core embedded in a non-absorbing shell (like water) – the core-shell particle will have much greater absorption than the core in isolation. However, the carbon core is still responsible for all of the absorption in a core-shell particle, as the water is not absorbing any photons.

Thus, the conceptual difficulty of attributing absorption to different species in an internal mixture should not be that difficult – we can compute the effect of any absorber in a model by removing it. For example, when we compute direct aerosol radiative effects, we compute the difference between "with aerosols" and "without aerosols." Likewise, direct aerosol forcing is computed by differencing the present-day aerosols with an estimate of the aerosol loading in some reference year (e.g., 1850). Why not do the same thing with individual aerosol species? Thus, the AAOD effect caused by a target species is: (total aerosol absorption) - (aerosol absorption in the absence of the target species). The resulting AAOD represents the physical impact of omitting the target species from the atmosphere, which is what we are seeking. Glancing through Table 2, it seems that CAM5-ATRAS is the only model that got this right (good job CAM5-ATRAS!).

Some of the AeroCom modelers have lamented in the past that this method of speciating absorption alters the aerosol size distribution, but so what? Our traditional computations of DARE and DARF dramatically alter the aerosol size distribution, too. In the case of DARE, after all, it totally removes the size distribution! Why is it folly to remove part of a size distribution instead of all of it? The other definitions in the table, including the old Aerocom protocol* recommendation at https://aerocom.met.no/protocol_expl.html, do not represent anything physical regarding internal mixtures and do not recognize that the absorption of an internal mixture is often greater than the sum of the absorbing components. In addition, the AeroCom protocol prescribes a set of densities that may or may not be consistent with any particular model. Thus, it seems as though this protocol was created mainly because of its simplicity.

*Aerocom protocol at https://aerocom.met.no/protocol_expl.html:
Aerosol optical depth per species for aerosol internal mixtures: A check on how you compute the contribution of a given aerosol species to total aerosol optical depth if you have internal mixtures in the model: Procedure recommended during AEROCOM workshop: Compute volume fraction of aerosol species in aerosol particle volume (without water!!) and retrieve with that fraction the aerosol optical depth for a given species. Apply the following densities for the major species (Dust = 2650 kg/m3 / Sea salt = 1600 / Sulfate = 1769 / Black Carbon = 1500 / Particulate organic matter = 1500)

Given the wide variety of methods for attributing AAOD to the absorbing species in this paper, the values for the component AAODs in Table 3 are not comparable between models. Since optical properties are computed offline, it would not be that difficult for each modeler to compute component AAODs with a single sensible protocol (ideally, as I described above for the internal mixtures) with a single set of refractive indices and densities for the absorbing aerosols. Constraining all of the models in the same way like this would make this paper strong and valuable. Otherwise, the paper just reports what we basically already know – the models are different from each other and the observations, and we have not quantified why.

Table 2

I really like the idea of Table 2 because it is a great idea to have the mixing assumptions for the AeroCom models all in one place. I don't understand some of the methods and terminology, though. For instance, what does *"Core-shell for internally-mixed BC particles; Volume mixing for pure BC and BC free particles"* mean for the CAM5-ATRAS model? A reader might surmise from this text that the authors are using volume-averaged refractive indices for external mixtures, which does not make sense.

It is also not clear which components are being internally mixed in many of the models. For example, ECHAM-HAM, ECHAM-SALSA, and GISS-MATRIX do not indicate which absorbing species they are treating as internal mixtures (BC, OA, dust, or some combination of all 3).

I was surprised to see that the EC-Earth3 model treats sulfate, ammonium-nitrate, organic aerosols, sea salt, and water as homogenous mixtures described by the Bruggeman mixing rule. This rule is usually applied to dry insoluble components, like mineral mixtures. The table seems to indicate that all of these components are mixed together into one internal mixture, but is that accurate?

The SPRINTARS model describes internal mixing of BC with OC in the 2nd column of Table 2, but the third column says that the BC AAOD is calculated assuming that all BC is externally mixed. Thus, this is inconsistent.

The NorESM2 model states:
Internal and external mixing. Maxwell-Garnett is used for calculation of refractive index of internal mixing of BC with other components, otherwise volume mixing.

What is volume mixing? If the modeling is applying volume averaging of refractive indices to external mixtures, then this is not the correct way to compute optical properties of external mixtures. Thus, "volume mixing" for external mixtures needs to be defined.

Figures

Figure 1 is based upon Mie calculations taken from *Samset et al.* (2018). However, *Samset et al.* (2018) provides information about the size distributions, refractive indices, and densities that are required to make this figure, whereas this article does not. Additionally, this article fails to explain that the *Bond and Bergstrom* (2006) recommended value of MAC = 7.5 m2/g can not be achieved using the *Bond and Bergstrom* (2006) recommended refractive indices and densities, which explains why the “fresh” symbols lie outside of the shaded region. Similarly, the “coated” and “collapsed, uncoated” symbols are left unexplained in this figure. *Samset et al.* (2018) explains that the symbols are based upon values found in the literature and lists several citations.

These are important details because a reader should be able to understand all figures on the basis of the material in the article. Additionally, since the caption states that MAC is separable amongst the species in Figure 1, the reader needs to know the range of size distributions and densities associated with the shaded regions.

Figure 2: Given that this paper is about modeling aerosol absorption, some discussion about the use of OPAC optical properties is warranted. Six of the 14 models in Figure 2 use BC imaginary refractive index (BC IRI) of 0.44, which is the value given by OPAC. That means that nearly 1/2 of the models are still using OPAC, but more than 1/2 of the models have figured out a way to move beyond OPAC. Way back in 2006, *Bond and Bergstrom* (2006) reported this in their assessment of BC refractive indices: “The value commonly used by climate modelers ($m = 1.74 - 0.44i$ at 550 nm) represents none of the possible refractive indices and should be retired.”

Bond and Bergstrom (2006) also said:
“The history of refractive index values tabulated by Shettle and Fenn (1976, 1979) is worth special mention. These are by far the most prevalent values for use in climate modeling, and have been incorporated into widely-cited literature, including a book by d’Almeida et al. (1991), and the Optical Properties of Aerosols and Clouds (OPAC) program (Hess et al. 1998). The original work by Shettle and Fenn (1976) averaged values from an earlier review by Twitty andWeinman (1971). In turn, the averaged data are taken from McCartney et al. (1965), who measured three coals, and Senftleben and Benedict (1918), who reported soot generated from an arc lamp. The review does not incorporate most of the findings on soot in the combustion literature, and indeed was written before most of that work was available. The precision of both n and k provided in OPAC values (three decimal places) is unwarranted, given this history. The OPAC value of $1.74 - 0.44i$ is drawn from incompletely graphitized carbon and has a lower value of k than most soot. . .
. . . The optical and physical data for LAC propagated by d’Almeida et al. (1991) have some interesting properties. Along with an imaginary refractive index that is too low, these authors recommend: (1) a particle size that is far too small (23 nm is the approximate size of primary spherules, not aggregates); (2) a geometric standard deviation that is somewhat too large (2.0);

and (3) a density that is far too low (a density of 1.0 is never observed; Fuller et al. (1999) tabulate measurements indicating densities of about 1.8 g/cm3). Despite returning to the string of citations that led to d'Almeida et al. (1991), we have been unable to unearth the sources of these values. When compared with measured values, each of the individual assumptions above may lead to an error of 50-75% in calculated properties that affect climate forcing."

It is unfortunate that so many models did not follow the *Bond and Bergstrom* (2006) recommendations. Since the present article is about model diversity of absorption, this would clearly be a great place to revive the OPAC BC issue.

Comparisons to AERONET data

Given the text on lines 192-200, does it even make sense to compare any extrinsic AERONET properties (like AAOD) to the modeled values, as in Figure 4?? That is, the AERONET dataset that the authors are using require $AOD(440) > 0.4$ (per line 193), but the models use all available AOD (per line 195). So why would there be an expectation that any modeled extrinsic parameter would compare well with AERONET under these conditions (even with a high-resolution grid)? If the authors checked the coincident-only AODs that are obtained during the same scans that produce the AAODs, I expect that they would see a significant bias in AOD for the same reason – modeled extrinsic parameters will not compare well to AERONET when you discard low-load cases in AERONET but keep low-load cases in the model output. (If they don't see a bias, that means that they are not using enough AERONET sites with $AOD(440) < 0.4$, or something is very wrong with the model). In fact, model comparisons to AERONET should be ***required*** to show coincident-AOD comparisons whenever there is a comparison of extrinsic parameters (like AAOD).

I suggest the following…plot aerosol co-albedo (absorption/extinction) instead of AAOD. Co-albedo is a pseudo-intrinsic parameter that is much less susceptible to column aerosol loading than AAOD.

Unclear Definitions for Organic Aerosol, Organic Carbon, and Brown Carbon

The authors are not clear about the differences between OA (organic aerosol), organic carbon, and brown carbon. Indeed, the text doesn't seem to recognize that BrC is a subset of OC and that OC is a subset of OA. On line 66, the authors refer to BrC/OA, implying that BrC and OA are one and the same species. On line 117 and in Table 2, though, the authors discuss OA/OC ratios, which are always greater than 1. On line 124, the authors mention that the *"OsloCTM3 model divide OA into into a mix of absorbing and non-absorbing species…"*.

The authors need to inform their readers that the brown shaded regions in Figure 1 correspond to organic aerosols that are "washed" with solvents like acetone and methanol in order to extract the absorbing organic aerosols from the non-absorbing organic aerosols. The OsloCTM3 model seems to recognize this by separating absorbing and non-absorbing OC. The other models apparently use a single set of optical properties that encompasses all organics (both absorbing and non-absorbing), so they should be using MACs that are much lower than the values shown in Figure 1. Additionally, BrC is produced by hulis and is largely found in biomass burning and is not observed in fossil fuel burning. Thus, the models should be using a different set of optical properties for OC that originate from modern urban areas than the optical properties that they use for biomass burning. None of these concepts are clearly recognized or discussed in the text.

Wavelength Dependence of Absorption

Section 3.4 and Figure 11 are another missed opportunity for some good discussion. All of the models except for OsloCTM3 show significant OA absorption at 870 nm, but Figure 1 indicates no detectable BrC absorption at this wavelength. Clearly, none of the models are using the recent measurements for BrC that are shown in Figure 1 (except perhaps OsloCTM3 – good job Oslo!). This begs the question – what are the models using for OA? Given that this is a paper on understanding the model spread of AAOD, this is an important topic. Sure, the authors provide OA refractive indices at 550 nm in Figure 2, but what measurements are the basis of the refractive indices in the models, and why are the modeled OA MACs and dust MACs buried in the supplement? The OA MACs in Fig S8 are on the low end of Fig 5 (assuming that the wavelength in S8 is 550 nm, but that is not stated), so does that mean that the OA imaginary refractive indices are more or less constant wrt wavelength? Also, the OA MAC varies by a factor of 4+ in Figure S8 – how come this huge range of modeled optical properties for OA is not discussed in the text? Same question for the MAC of dust, which varies by a factor of 3+ in Figure S8.

Going Forward

There is potential for an excellent paper in this material. I would like to see:

- An analysis of the models with external mixing. What does the spread in MACs and AAODs for this subset of models look like, and what is the cause of the spread? Size, refractive index, and density are the important parameters. Analyzing MAC wrt IRI/density will probably be enlightening. Column mass loading of the absorbing aerosols should be a factor.
- A single optics module applied to the models with internal mixing, as much as possible. Use the CAM5-ATRAS method for splitting absorption amongst components and analyze the spread in MACs and AAODs, and abandon the old AeroCom protocol.

Pay attention to the mass/volume fraction of absorbers in the mixtures, especially for BC. Here again, column mass loading will be important for understanding AAOD.

- With separate analyses of the external and internal mixtures now in hand, how do these two analyses compare to each other? How do they compare to the models with internal mixing that are not amenable to the approach in the 2nd bullet?
- Don't compare to any extrinsic AERONET parameters, since the $AOT(440) > 0.4$ restriction skews the AERONET data to large aerosol loadings. Use AERONET's single-scatter co-albedo, or omit.
- Pay attention to BC density. The use of OPAC BC with a realistic density (as opposed to OPAC's recommended value of 1 m2/g) is probably the cause of the lowest MACs in Figure 9. This could be easily remedied by abandoning OPAC. The densities of all absorbers should be included in a table.
- Abandon OPAC. Since the authors are only considering a few wavelengths (440, 550, and 870 nm), abandoning OPAC is not a tall order.
- Inform the reader about where each model obtains their information for optical properties of aerosols (i.e., provide citations). Many of them use OPAC, but where do the other models find complex refractive index, size, and density information? This will allow the reader to understand which models are using the latest measurements, and which ones are lagging.
- A description of the size distributions that the models use is needed. Lognormal radii, widths, etc., should be tabulated.
- Make sure that the figures are self-contained (in the sense that the reader does not have to go to another paper to understand how they were created).
- Clear up the phraseology everywhere, especially in Table 2 where it is so important. Make sure that all text is clear so that a reader understands the intent of your words without ambiguity. There are many co-authors on this paper for proofreading, so this should not be an issue.
- Clearly define OA, OC, and BrC early in the text.

All of this should be do-able with the model runs already utilized in this paper.

Line-by-line issues that need to be addressed:

Line 70:
"While fine-dust particles mostly scatter solar radiation, coarse dust also absorbs moderately in the visible and near-infra-red spectrum."

I would like to see a citation for this, because I am not sure if it is accurate. Iron oxides in dust are small, so I don't see why they would not be present in the fine-mode dust as well as coarse mode dust.

Line 157:
"…their imaginary parts of the refractive index vary a lot ($1.75 + 0.44i$ *for SPRINTARS and* $1.85 + 0.71i$ *for GISS-MATRIX (Fig. 2)."*

This is a missed opportunity – why not use the same BC refractive index and BC density in all of the models? This would eliminate a significant source of diversity so that part of the analysis could be focused on the remaining causes of diversity (i.e., size distribution and column mass loading). Additionally, the diversity of BC density is entirely missing from this paper. BC density is very important because it is inversely proportional to MAC (all else being equal), and therefore has a direct impact on both MAC and the AAOD computed by the models. Finally, why not compare column mass loading amongst the models as well? I believe that we could learn something by studying mass diversity alongside the MAC, AOD, and AAOD diversity, but column mass loading was not even mentioned as a source of diversity in this article.

Line 199:
"However, using a high-resolution simulation of global aerosols, Schutgens (2020) found a much smaller bias of 9%"

Ending the paragraph of caveats with this sentence is a bit of a hoodwink, as the present study does not use a high-resolution model.

Line 213:
"In NorESM2 the additional absorption is from sea-salt and sulfate (mixed with BC, dust and OA). In GFDL BC is internally mixed with SO4, so the additional absorption is due to SO4 (mixed with BC)…"

This is not explained very well because the most important part of the explanation is in parentheses. Reading this, one might think that sea-salt, sulfate, etc. are absorbing photons. The authors should clearly point out that internally mixed BC has greater absorption than externally mixed BC, and that this is because the internal mixtures have greater geometrical cross-sectional areas than the BC inclusions within the mixtures. The sea-salt and sulfate in the mixtures are not absorbing any photons, whether they are hosting absorbing inclusions or not. This needs to be clear.

Line 323:
The authors mention BC particle size as 50 nm, but they do not state whether this is radius or diameter. Additionally, 50 nm is closer to the diameter of a spherule than a BC aggregate. For example, *Schwarz et al.* (2008) measured size distributions of BC with an SP2, and they obtained a median diameter of ∼200 nm.

Line 363: If the global mean modeled AOD is 0.129 and the global model mean AAOD = 0.035 (per line 188), then the global mean SSA should be close to (1 - .035/.129) = 0.73. Why is the global mean AAOD so high wrt to the global mean AOD?

Line 391-393:
"We also find very little correlation between the imaginary index and mass absorption coefficients. For BC just three different refractive indices are used by the models, while the spread is not related to this choice."

This sentence appears in the Summary, but I don't see IRI/MAC correlations discussed in the paper. However, it is not surprising that the authors do not see a relationship between IRI and MAC if they did not account for the different densities used in the models. For instance, OPAC uses a density of 1 g/cm3, which results in a MAC of ∼10m2/g (per *Bond and Bergstrom*, 2006). A more realistic choice of 1.8 g/cm3 for BC density would reduce the MAC to 10/1.8 = 5.5 m2/g. The imaginary refractive index makes a huge difference to the BC MAC, but not if one simultaneously tunes the BC density.

Line 398:
"We recommend that the role of size and mixing rules and diagnostic procedures should be investigated in more detail to understand the differences in mass absorption coefficients."

I agree completely, but why didn't the authors do that in this paper? That is actually what many readers will be looking for in this article.

Line 403:
"In particular, we have found that the imaginary indices are not explaining much of the AAOD variance, except slightly for dust."

Here again, I did not see this discussed at all in the paper. The final section should be a summary of the details that are presented in the paper, not the introduction of a new result.

Minor issues:
Line 51:
"The three absorbing species are rarely observed as single species,..."
This sentence does not make sense to me... Shouldn't this go unstated by definition?

Line 58:
"but these calculations are approximate (using mixing rules or the assumptions of a co-centric core/shell structure)..."
This sentence seems to imply that internal mixing requires additional significant assumptions wrt external mixing, which is not the case. Core/shell computations are exact, although the core/shell structure is an approximation for the shape of aerosols in the atmosphere. Likewise, Mie Theory is also exact for the spherical particle approximation that is used for all externally-mixed aerosols in the models (with the exception that some models use a spheroidal approximations for dust), but spheres and spheroids are still approximations for

particle shape. Thus, approximations associated with shape are required for both internal and external mixing. Finally, errors associated with effective medium approximations have been tested by many authors (e.g., *Martins et al.*, 1998; *Fuller et al.*, 1999; *Lesins et al.*, 2002) and are likely swamped by other modeling errors associated with mixing fractions and assumed component refractive indices.

Line 61: Begins with "However,…" and then essentially repeats the information on line 58. Sentence should start with a different word.

Line 102: "the separation into fine ($< 1\mu$m) and coarse mode ($> 1\mu$m) AOD…" Presumably the authors mean diameter, but this should be specified.

Line 119: "and 11 models have provided absorption split into BC and dust (OA)." Why is OA in parentheses?

Line 147: Authors are using Version 2 AERONET, but which level? That is, Level 1.5, or 2.0?

Line 183 (Fig 4) and Line 185: The authors need to specify whether they are using Version 2 or Version 3 AERONET and Level 1.5 or Level 2, here. The authors mention Level 2 later on Line 193, but it is not clear whether they realize that Version 2 means something different than Level 2.

Line 228: Table 2 says that Sprintars has internal mixtures of BC with OA. Why do the authors include it as a model with external mixing?

Line 276: Replace "Methods" with "Section 2 – Methods" for improved reader navigation.

Line 325 & 328: BC AAE was addressed in *Schuster et al.* (2016b), not *Schuster et al.* (2016a).

Line 335: Should be Fig 13, not Fig 10.

Line 360:
"Compared to retrieved AAOD from AERONET stations, the models yield lower absorption."

One would hope so, since the AERONET dataset that the authors use requires $AOD(440) > 0.4$, whereas the models are not using the same restriction.

Line 381: It is good that that the authors are recommending and update for modeled OA, but it would be nice if they reported what the models are actually using right now, too.

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