Review of paper:

A new MISR research aerosol retrieval algorithm: a multi-angle, multi-spectral, bounded-variable least square retrieval of particles properties over both land and water validation *by J. Limbacher et al.*

Highlights

- very detailed MISR retrieval approach of the research algorithm (RA)
- evaluations not just limited to AOD
- nice illustrations of strength and weaknesses

Concerns

- comparisons of results to the older RA and also to the standard (SA) algorithm
- likely incorrect assumptions about dust bias solutions
- regional (Sahara, off-Sahara) testbed cases are missing to examine the dust problem

General comments

MISR comprises a set of multi-spectral sensors oriented into different directions. Data combination in developed retrievals are quite powerful to determine multiple aerosol properties (at cloud-free conditions) with accuracies usually not matched by other satellite sensors. In addition, the long-term data record (in operation since 2000) makes this data-set highly attractive.

While a new retrieval is suggested, the 'research' aspect makes me frown, as new efforts for retrievals will only count for data-users, if retrievals are applied to the entire data-record (e.g. for climate studies) and not to a limited number of cases. In any case, statistical comparisons of retrievals (newRA vs oldRA vs stdR) for a limited period could be a nice addition to demonstrate newRA capabilities - in the discussion section.

The paper is rather technical and introduces a new (less complex) aerosol model with the number of permitted aerosol types – compared to the standard algorithm - reduced to 17. This is a step in the right direction (also for unique answers). Still I suggest further changes. I question the necessity of 'very small' (reff ~0.06um) sizes, which are hardly contributing to optics – unless they are very absorbing. However, 'very small' BC is quickly increased in size to 'small' and even to 'median' sizes as (absorbing OC and scattering SU) condensate attaches. Hereby for OC a weak absorption in the mid-vis but a strong absorption towards the UV should be assumed so that a BC(core, reff~0.06um))/OC(shell) type can mimic 'brown carbon'. Thus, there is high potential to reduce the number fine-mode choices. On the other hand, the coarse-mode choices are far too simple as also larger mineral dust sizes (with lower mid-vid SSA for the same Rfimag) should be considered. I am almost certain, that this will reduce FMF, non-sph and ANG biases of this new MISR retrieval.

Otherwise, this is a very informative paper.

P.S. I have attached a summary of the top-down concepts of my MACv3 climatology, where is the coarse-mode AAOD information – along with the dust coarse-mode AOD is used to determine coarse dust AOD and coarse dust size.
(note, fine-mode dust AOD and fine-mode SSA contributions are considered secondary and being considered conservative scatters, they are attributed to non-absorbing fine-mode 'SU' in the MAC climatology)

Specific comments

5/4 to table 1: I missed a couple of (at least one) larger dust sizes (as with larger dust AOD usually also the mineral dust sizes are larger … which strongly adds [coarse mode] absorption). I also would get rid of the very small aerosol types and would start with 'small'. Here I would add a mixture (a 'very small' BC core size with an organic OC shell to yield a 'small' mix type) as mostly (or only) scattering usually quickly condenses on BC. Hereby I would also define organic ('OC') with a strong increasing absorption increase towards the UV and a pure scattering fine-mode ('SU') component for both 'small' and 'medium'). Hereby the BC(core)/OC(shell) type covers the artifical 'brown carbon' component. This reduces the minimum number of types to be considered to eight: small: BC/OC, SU, OC / medium: OC, SU / large: SS and DU / very large: DU
For the content in the table I would like to see next to the eff. radius also assumed distribution width information (std.dev or variance) rather than r,min and r,max. In addition, for the Angstrom parameter, the defining wavelengths need to be listed in the captions and SSA data should be shown that actually relate to the type assumption for size and composition (and not just made up by an arbitrary value, like 0.8). This will also help later to improve to relate types to those used in global modeling. Finally for AAE (as fro Angstrom) the defining wavelengths are needed or simply add an SSA value at another relevant wavelength (e.g. 440nm) – based on RFimag spectral data for the particular size of that type.

6/6 one large non-spherical model is not sufficient - especially over the Sahara and for Saharan outflow - where extra large dust-sizes, if ignored, likely cause AODc underestimates and also possibly absorbing fine-mode overestimates

7/10 why not using initial values from a climatology or data from the most recent retrieval at that location?

9/7 actually it would be great if this mixture information could be saved – at least for a couple of test-cases. In this way I could be explored to what degree each of the now 17 types contributes also in efforts to reduce the number of required types.

9/25 if you remove the smallest (re=0.06um) sizes (then you are down from 9 to 7) and when you add a larger non-sph dust (e.g. re ~ 5um) then you are up from seven to 8)

11/12 I assume that in the combined surface/aerosol retrieval constraints are built in, which do not allow for negative albedos or negative AOD.

18/24 Nice, that the comparison are shown for both the largest and (via ln/ln) the smaller AODs

20 I think, that the missing coarse mode (e.g. MISR Angstrom overestimates at smaller AERONET Angstrom) has much to do with the missing larger coarse-mode sizes. This would have been also apparent, if the deviations of the scatter plot would have been places as a function of location. (This behavior is similar to SLSTR biases, which also only consider one dust size in their model and then attribute absorption to the fine mode (AODf overestimates), while this absorption should have gone to dust size (increases))

22/10 For me the fine-mode non-spherical dust is not the issue. If absorption is allowed not only to be associated with fine mode (BC, OC) and relative small DU, but also with larger DU size, then most of the MISR biases now will go away (better non-sphere, better fine-mode, better SSA) in comparisons to DUST cases (on the other hand at the largest AODs AERONET actually gives size-distributions, although size with re>10um are likely missed by these inversions). In the scatter plot presentation the 0.5-1.0 AOD already relates to the largest AOD events, so I consider only the first row in figures 5 and 9 relevant. (rows 2 and 3 in Figure 5 and row 3 in Figure 9 are interesting but less meaningful because of much lower statistics are less meaningful). Why not showing in Figure 5 the same range statistics as in Figure 9?

29/4 AOD >0.4 is already a large AOD and AOD>1 are very rare, so I suggest to focus on the 0.3 to 1.0 range for coarse mode AOD. And the scatter plots for FMF, non-sph and SSA do not indicate co-locations … so I would look at dust outflow off Africa (ocean) and dust over the Sahara (land) to investigate the dust retrieval problem.

Attached, below is the description of the top-down approach of the MAC aerosol climatology. In the top-down approach also the dust size is retrieved (from the coarse-mode AAOD). In the third column for Figure A3 the extracted dust effective radius (divided by 10) is illustrated. Especially over the Sahara and for dust outflow ontoe Atlantic – especially during JJA due dust size are significantly larger … even for monthly averages (of this climatology).

MAC v3 details

The Max-Planck Aerosol Climatology (MAC) offers merged monthly global maps for aerosol optical properties. In the merging process, multi-annual observational statistics of photometry from the ground is forced on spatial context supplied aerosol component 'bottom-up' modeling. The merged aerosol optical properties focus on aerosol column amount and aerosol column absorption, separately for smaller 'fine-mode' aerosol and larger 'coarse-mode' aerosols. In Figure A1, multi-annual averages from photometry (AERONET/MAN with absorption data only over continents) are compared to multi- (AeroCom phase 3) model interquartile averages (AC3-iqa). In addition, in that Figure A1 the resulting MAC version 3 maps are presented along with applied regional % changes to AC3-iqa.

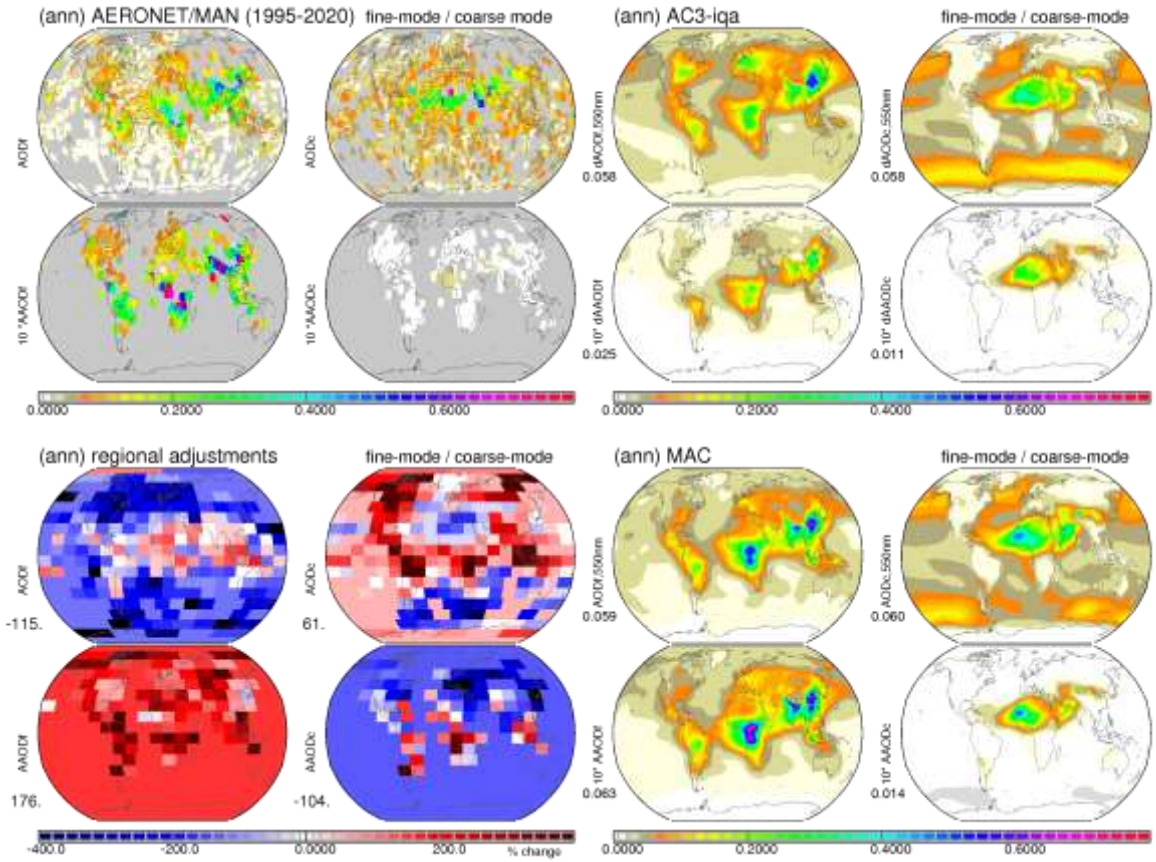

Figure A1. *Global annual average distributions for AODf, AAODf, AODc and AAODc of (1) AERONET/MAN photometry (upper left), (2) the AeroCom phase III multi-model interquartile average background (AC3-iqa, upper right) and (3) resulting MAC climatology fields (lower right) - after AERONET/MAN adjustments in % to AC3-iqa (lower left). Numbers at the lower left of each image indicate global averages. Also note that absolute aerosol absorption data (AAOD) are multiplied by 10 to match the common scale.*

Major adjustments to the 'bottom-up' modeling background are strong increases to the 'fine-mode' absorption (AAODf) and decreases to the 'fine-mode' aerosol amount (AODf) at mid-latitudes. Also SH continental dust (AODc) is stronger. Absolute MAC version 3 changes with respect to both

background (AC3-iqa) and to the older version2 of the MAC climatology (*Kinne, 2019a*) are presented in Figures A2 and A3.

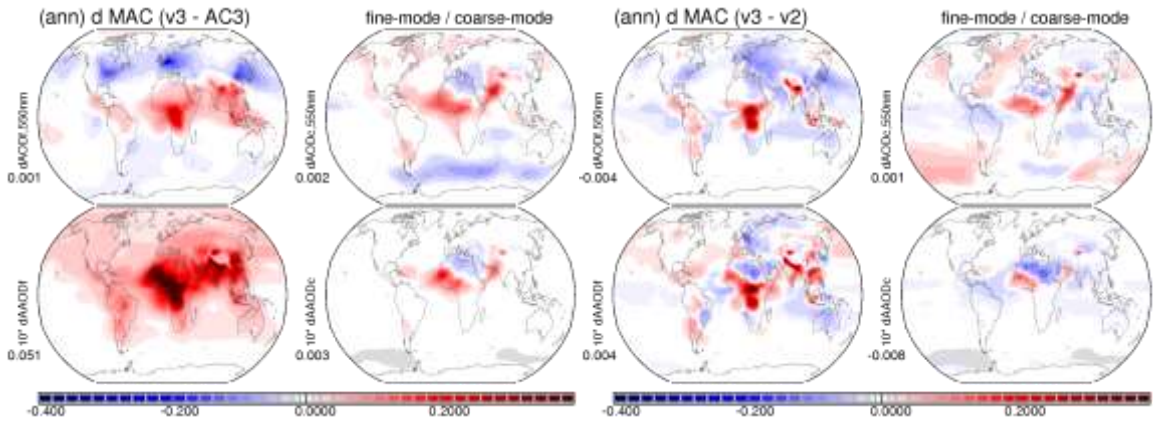

Figure A2. *Absolute annual difference global distributions for AODf, AAODf, AODc and AAODc between the MAC (v3) climatology and the AC3-iqa background (left block) or the older version (v2) of MAC (right block). Average differences are summarized by a value and for the common scale absorption data differences were multiplied by 10.*

Compared to the previous MAC (v2) version (*Kinne 2019a*), the new MAC (v3) climatology version uses and more recent (better emission data applying and component mixture permitting) modeling AC3-iqa background (instead of the AC1-median) and considers 238 (instead of 21) sub-regions for regional adjustments. This resulted in different fine-mode distributions over E. Asia on strongly increased fine-mode contributions over central Africa.

In a 'top-down' approach aerosol column and absorption properties are associated with a mixture of pre-defined aerosol components, that differ in size and mid-visible absorption. All MACv3 types are listed in Table 1.

Table 1. *size and absorption potential of pre-defined aerosol types in MACv3. Listed are (1) effective radii (Re) of log-normal distributions with associated mode radii (Rm) and standard deviations (sd), (2) mid-visible refractive indices (and from size and RF,imag resulting SSA values) and (3) column numbers (N) based on component global mid-visible AOD averages. Five different sizes are allowed for sulfate and dust, each. For comparison, values of a cumulus water cloud (water) and a cirrus cloud (ice) are provided.*

aerosol type	label	Re	Rm	s_d	RF_R	RF_I	SSA	< OD >	N
		[um]	*[um]*		*at 550nm wavelength*				*[#/m2]*
soot (not used)	BC	.06	.03	1.7	1.70	.7000	.155	0.005	4.0 e+11
soot + o.shell	**BO**	**.12**	**.08**	**1.5**			**.615**	**0.015**	**4.0 e+11**
organic	**OC**	**.18**	**.12**	**1.5**	**1.53**	**.0050**	**.975**	**0.022**	**1.8 e+11**
sulfate	SU	.06	.03	1.7	1.43	.0000	.999	0.023	4.4 e+13
sulfate	SU	.10	.05	1.7	1.43	.0000	.999	0.023	4.1 e+12
sulfate	**SU**	**.16**	**.08**	**1.7**	**1.43**	**.0000**	**.999**	**0.023**	**6.0 e+11**
sulfate	SU	.26	.13	1.7	1.43	.0000	.999	0.023	1.2 e+11
sulfate	SU	.40	.20	1.7	1.43	.0000	.999	0.023	3.8 e+10

seasalt	**SS**	**2.5**	**.75**	**2.0**	**1.50**	**.0000**	**.999**	**0.035**	**3.3 e+09**
dust	**DU**	**1.5**	**0.93**	**1.55**	**1.53**	**.0011**	**.962**	**0.025**	**2.7 e+09**
dust	DU	2.5	1.34	1.7	1.53	.0011	.962	0.025	1.3 e+09
dust	DU	4.0	1.55	1.85	1.53	.0011	.962	0.025	7.0 e+08
dust	DU	6.5	1.98	2.00	1.53	.0011	.962	0.025	3.6 e+08
dust	DU	10	2.30	2.15	1.53	.0011	.962	0.025	2.0 e+08
cloud **water**	water	10	6.7	1.5	1.33	.0000	.999	10.0	2.5 e+10
cloud **ice**	Ice	40	20	1.7	1.31	.0000	.999	0.5	1.1 e+08

For smaller fine-mode sizes (1) a strongly absorbing BO (a soot coated by organic material) type, (2) a weakly absorbing organic matter (OC) type and (3) a non-absorbing sulfate (SU) type are considered. Thus, the SU type includes other non-absorbing fine-mode contributions, as from nitrate or seasalt. In order, to separate absorption contributions between BO and OM, the BO/(BO+OM) ratios for AOD from AeroCom 'bottom-up' modeling are applied (i.e. BO/(BO+OM) AOD ratios are higher over urban pollution than over wildfire regions). While BO (Re=0.12um, with Re=0.06um BC cores) and OM (Re=0.18um) sizes are fixed, the SU size (0.06 < Re < 0.40um) remains variable to match fine-mode effective radii of the MAC climatology.

For larger coarse-mode sizes (1) a non-absorbing seasalt (SS) type and (2) an absorbing mineral dust (DU) type, are assumed. While the SS (Re=2.5um) size is fixed, the DU size (1.5 < Re <10um) is assumed to increase with coarse-mode DU-AOD. Note that even with a constant mid-visible imaginary part for dust – here assumed at 0.0011 (*Di Biagio, 2019*) – the mid-visible absorption potential ($1\text{-}SSA_{DU}$) increases with dust size. With an initial guess for the SS-AOD to extract the AODc associated with mineral dust, the following relationship is applied:

$$1\text{-}SSA_{DU} = 1\text{-}SSA_{DU,min} + 0.05 * \text{DU-AOD}$$

$1\text{-}SSA_{DU,min}$ = 0.037 (for the smallest assumed dust aerosol radius of 1.5um)
DU-AOD = (AODc – SS-AOD,guess)
SS-AOD,guess = $.003 * \text{windspeed}_{SUR}$ [m/s] *(2-cos (2* (lat[deg]-sun[deg])/2)
*(ocean_fraction)

As the coarse-mode absorption now defines both the DU-AOD and the size for dust, and the remaining coarse-mode AOD is assigned to seasalt to replace the initial SS-AOD guess.

Figure A3 presents seasonal averages for 'top-down' size-choices for sulfate and dust components, for the fine-mode Re of MAC and for the applied BO/(BO+OM) ratios from modeling.

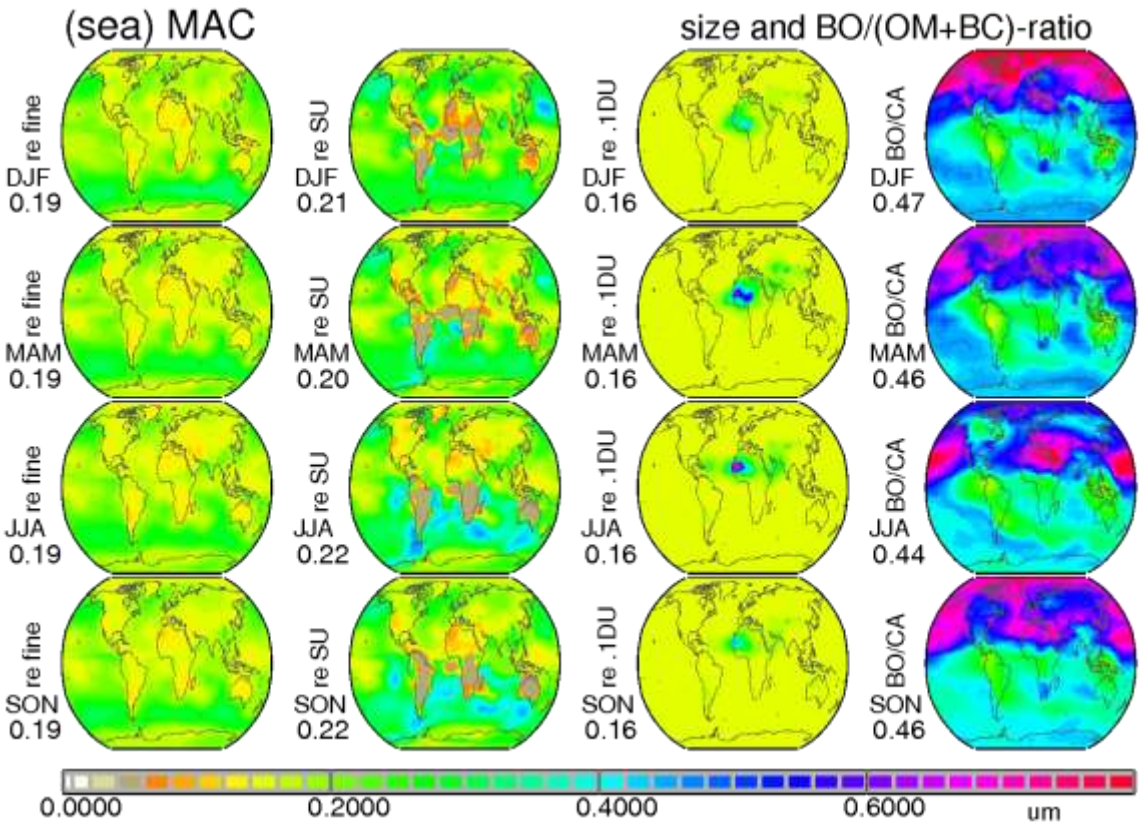

Figure A3. *MACv3 seasonal maps for fine-mode Re (col1) and as part of the 'top-down' approach sizes (Re) for non-absorbing fine-mode (col2) and mineral dust (col3 – times 0.1, to fit common scale). Also presented are applied BO/(BO+OM) ratios from modeling (col4) to separate BO and OM. Numbers next to plots show seasonal averages.*

The resulting aerosol component AOD maps attributed in the 'top-down' approach are presented Figure A4: seasalt (SS) and dust (DU) from AODc and sulfate (SU), organic carbon (OC) and black carbon from AODf.

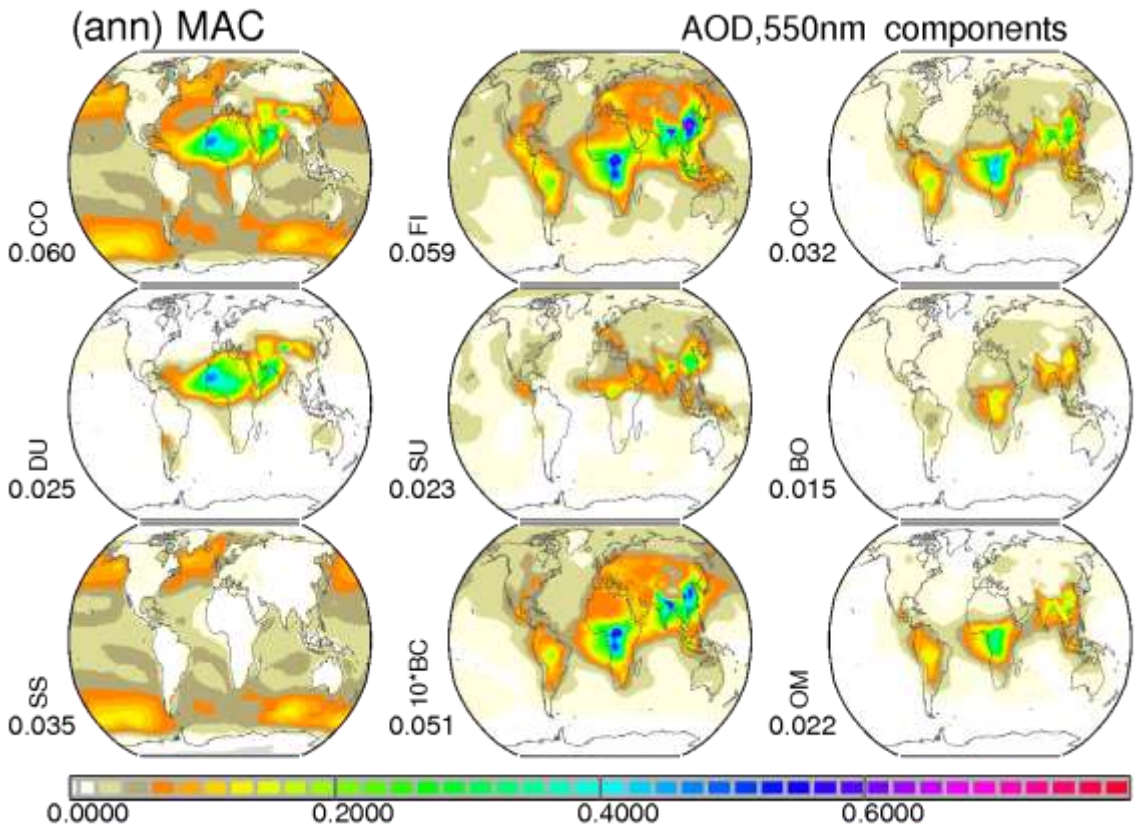

Figure A4. *MACv3 annual maps of 'top-down' derived AOD component distributions. The coarse mode AOD is split between seasalt (SS) and mineral dust (DU) contributions (left column). The fine-mode AOD is separated into non-absorbing sulfate (SU), strongly absorbing BC (multiplied by 10) and weakly absorbing OC. In a different split for total carbon (CA =OC+BC) contributions without soot (OM) and contributions containing a soot core (BO) are separated. Values next to the maps indicate global averages.*

The annual maps of Figure A4 are a subset of maps shown in Figure 2. To illustrate MAC updates with this version 3 annual difference maps to the previous version 2 (Kinne, 2019a) maps are presented in Figure A5. Global coarse-mode AOD contributions remained stable but DU-AOD is smaller (as well as maximum DU sizes) and SS-AOD is larger. Reduced global fine-mode AOD contributions are caused by significant SU-AOD reductions despite increases to OC-AOD and BC-AOD. Major regional component AOD differences between v 3 and v 2 of MAC are:

DU	- 0.005	more DU over Arab waters, W. Africa, Patagonia, less DU over N. S. Am. and N. Africa
SS	+0.007	more SS over mid-latitude oceans
SU	- 0.015	more SU over N. India and central Africa, less SU over E. Asia, E. Europe, E. US, tropics
OC	+0.010	more OC over central Africa, N. India, SE. Asia, S. America
BC	+0.001	more BC over central Africa, N. India, northern hemisphere

As the fine-mode AOD is smaller, also the anthropogenic (fine-mode) AOD is smaller and less absorbing with the sharp reduction to (scattering) SU-AOD. Also, the anthropogenic (coarse-mode) dust AOD is smaller.

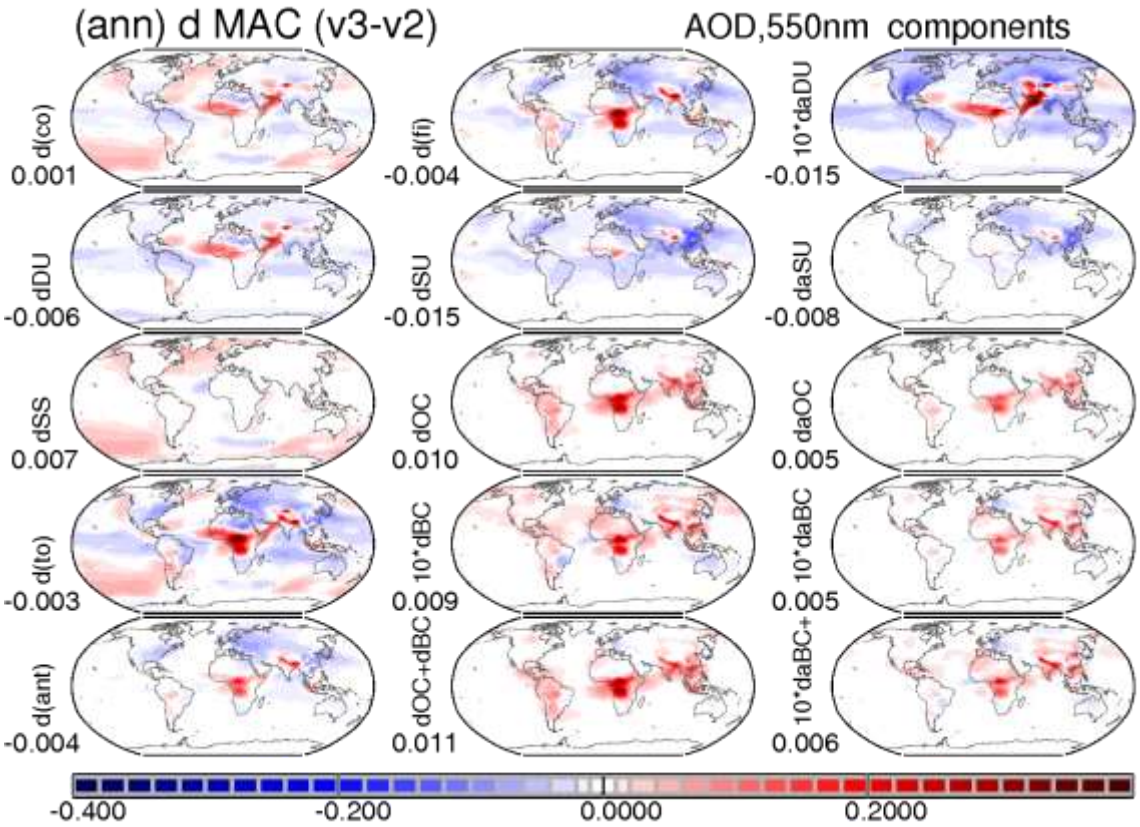

Figure A5. *Absolute annual differences for 'top-down' component AOD data between the current MAC version3 (see Figure 2) and the previous version 2. Values next to the plots indicate global average differences.*

The big advantage of separating AODc and AODf in aerosol components which are completely defined by size (-distribution), composition (with its known refractive indices over the entire spectral solar and infrared spectral region) and shape (here spheres are assumed) is, that all three spectrally varying single scattering properties (as input for broadband radiative transfer simulations) are quickly calculated. This is done via (MIE-) scattering simulations for (1) extinction (EXT, attenuation per distance), (2) single scattering albedo (SSA, the scattering potential) and (3) asymmetry-factor (ASY, approximating the scattering distribution). These single scattering simulations can be done for every desired spectral (radiative transfer) model resolution, as long as the component refractive indices at that resolution are provided. The presented spectral (8 solar and 12 infrared) choices in Figures A6 and A7 below refer to the spectral resolution of the a subsequently used radiative transfer model. The addressed aerosol components in Figure A6 and A7, which were already introduced in Table 1, are soot (BC, Re=0.06um), an organic mixture with a soot core (BO, Re=0.12um), organic material (OC, Re=0.18um), sulfate (SU, Re=0.16um), seasalt (SS, Re=2.5um) and mineral dust (DU, Re=1.5). In addition, properties of a larger mineral dust size (DU+, Re=6.5um) and for a general comparisons also properties for a water cloud (water, Re=10um) and for an ice-cloud (ice, Re=40um) are included. In Figure A6, size-distributions and component refractive indices

are compared. Note, that for the component shell/core mixture of the BO component no combined refractive index is offered (internally calculated) and that for mineral dust and sulfate, independent of a selected aerosol size, the same refractive indices apply.

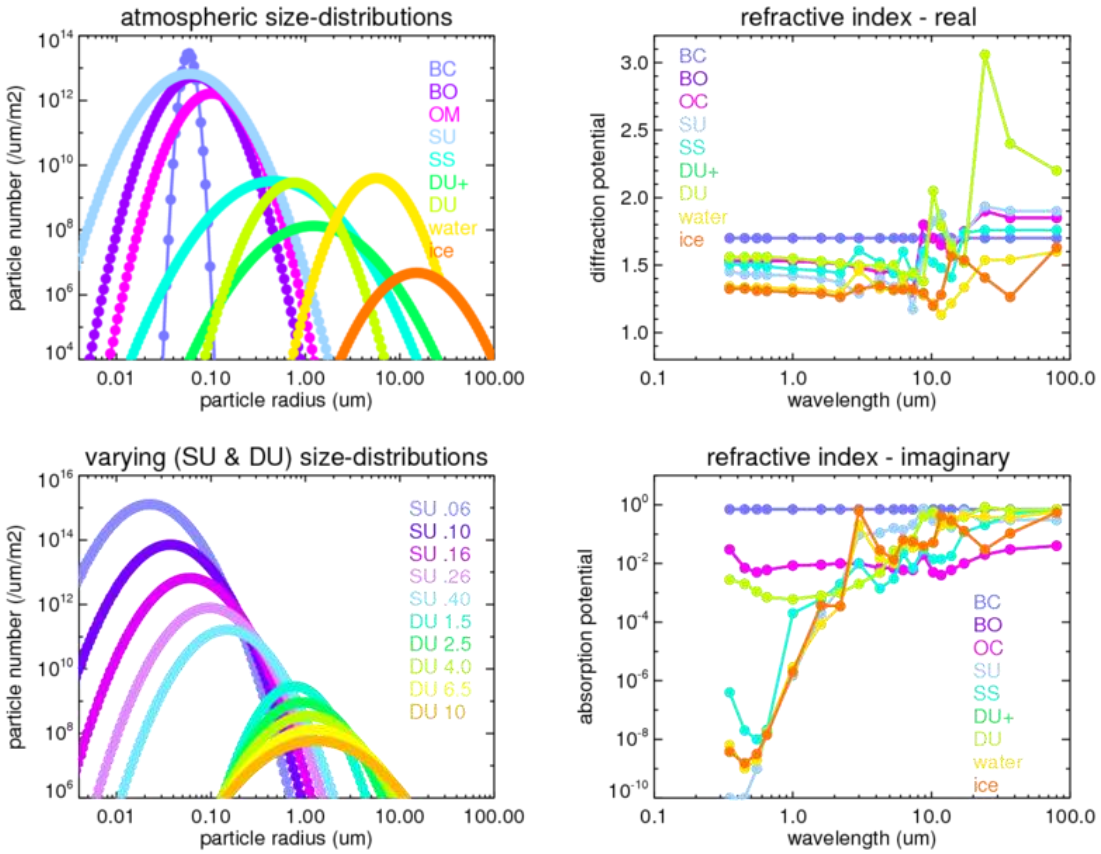

Figure A6. *Aerosol size distributions (left images) for pre-defined aerosol components (to match in presented concentrations the global average MACv3 AOD) and real and imaginary parts of the refractive indices (right images). Refractive indices are compared at central wavelengths of 8 solar and 12 infrared spectral bands. For comparisons, the size distribution and refractive indices for a cumulus cloud and for a cirrostratus cloud are shown.*

Mie simulation (assuming spherical aerosol shapes) then yield the single scattering properties (EXT, SSA, ASY), which are presented for the components of Figure A6 in Figure A7.

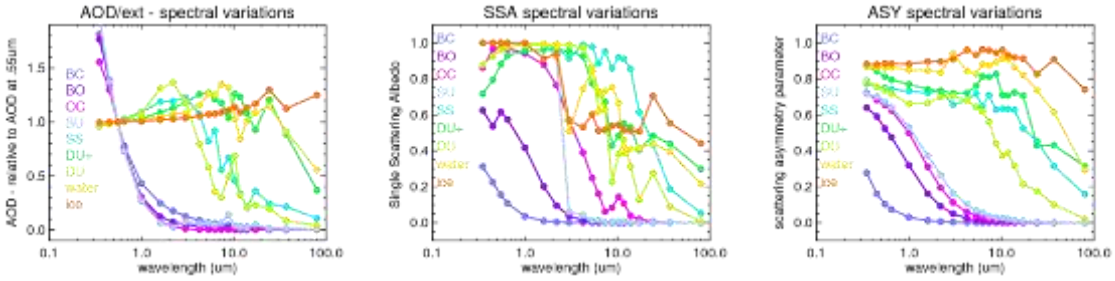

Figure A7. *Calculated spectrally varying component single scattering properties for extinction (left - via the ratios to the component extinction at 550nm), for single scattering albedo (center) and for the asymmetry factor (right).*

According to AOD maps of Figure A4, component single scattering properties are combined (AOD is additive, SSA is weighted by AOD and ASY is weighted by AOD*SSA) to yield global maps. Resulting single scattering properties at four selected wavelengths for fine-mode and coarse mode aerosol are presented in Figure A8.

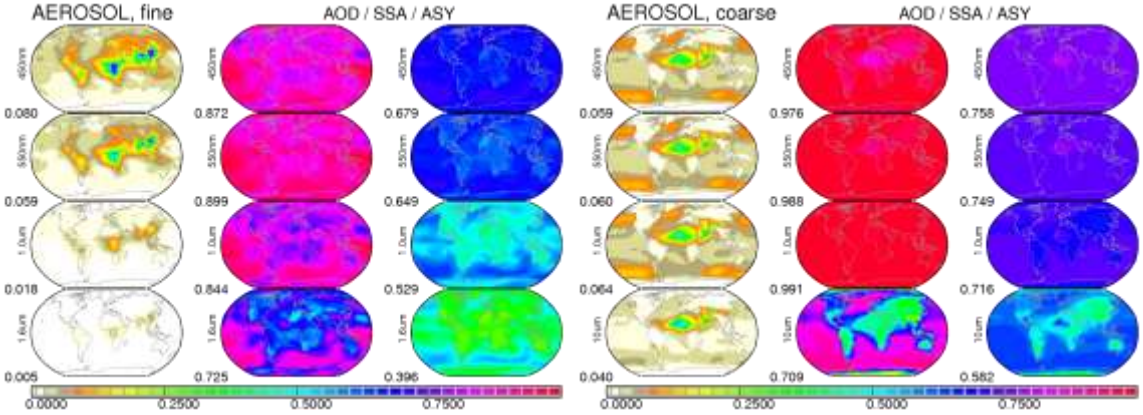

Figure A8. *Component combined annual average single scattering properties maps (AOD – left col., SSA – center col., ASY – right col.) at four selected wavelengths: at .45, .55, 1.0 and 1.6um for the fine-mode AOD (left) and at .45, .55, 1um and 10um for the coarse-mode AOD (right). Numbers at the lower left indicate global annual averages.*

Note that for the fine-mode, the aerosol sizes are too small to yield significant radiative infrared effects (at wavelengths >4um), so that data are presented at another near-IR wavelength (1.6um - rather than 10um). Similarly to Figure A8, the resulting single scattering properties at four selected wavelengths for (fine-mode) anthropogenic aerosol and for total (fine-mode and coarse-mode combined) aerosol are presented in Figure A9.

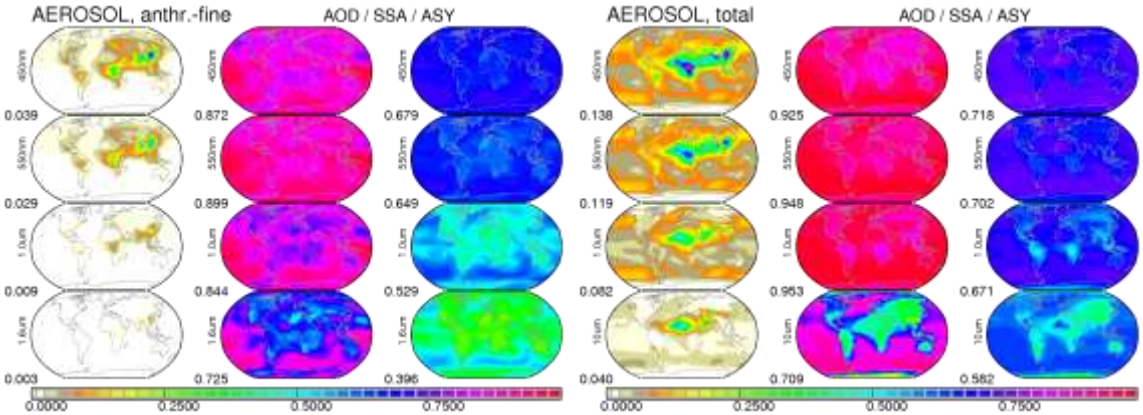

Figure A9. *Component combined annual average single scattering properties maps (AOD – left col., SSA – center col., ASY – right col.) at four selected wavelengths: at .45, .55, 1.0 and 1.6um for anthropogenic*

(fine-mode) AOD (left) and at .45, .55, 1um and 10um for the total AOD (right). Lower left numbers indicate global annual averages.

Now all aerosol needed optical properties are defined, so that radiative transfer simulation can be performed to determine the aerosol radiative effects.