Harmonization of aerosol approaches in remote sensing and transport models

Atmosphere Monitoring

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4 – HYGEOS, Lille, France,   *5 – ECMWF, Reading, UK*

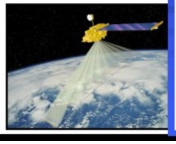
GRASP HYGEOS

Laboratoire d'Optique Atmosphérique   EUMETSAT   ECMWF

Motivation:

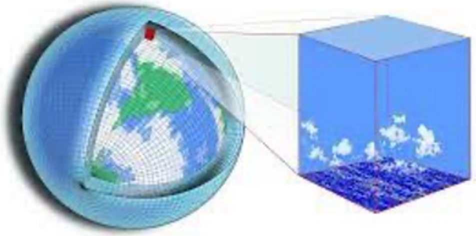
Remote sensing:

- passive
- active
- satellite
- ground-based
- airborne
- etc.



Transport Modeling / Reanalysis:

Spatial and temporal distributions of aerosol microphysical properties



aerosol assumptions are

= or ≠

- **Gap** between aerosol modelling approaches used in different remote sensing algorithms and in the global climate models.

CAMS

MERRA-2

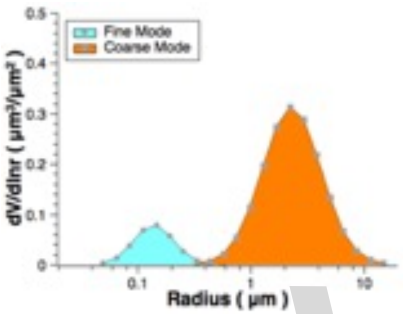
MERRA-2 Natural Run

Aerosol components and bins (tracers)

	Aerosol		CAMS	MERRA-2	MERRA-2 Natural Run
1	**BC**	Hydrophobic	X	X	X
		Hydrophilic	- (X: in new cycle)	X	X
2	**OM**	Hydrophobic	X	X	X
		Hydrophilic	X	X	X
3	**SU Hydrophilic**		X	X	X
4	**Sea Salt Hydrophilic**	SeaSalt1	X	X	X
		SeaSalt2	X	X	X
		SeaSalt3	X	X	X
		SeaSalt	-	X	X
		SeaSalt5	-	X	X
5	**Dust**	Dust1	X	X	X
		Dust2	X	X	X
		Dust3	X	X	X
		Dust4	-	X	X
		Dust5	-	X	X

Aerosol model in GRASP:

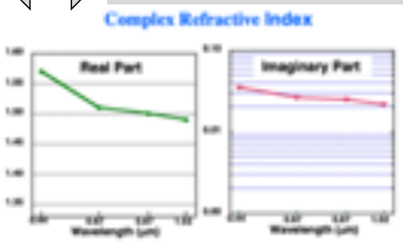
Multi-component mixture of spheres and randomly oriented spheroids



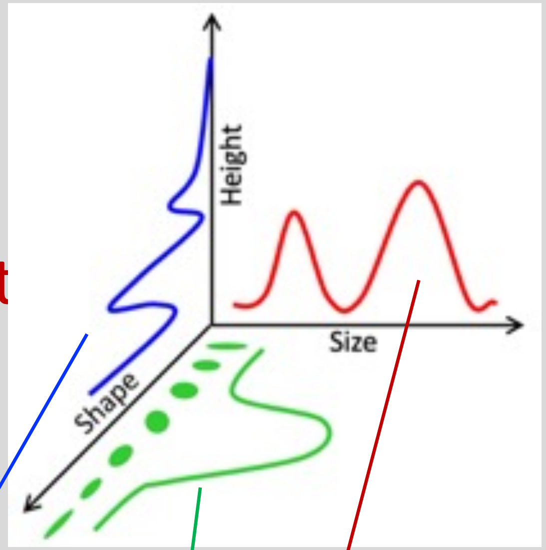
TO be RETRIEVED
OR
ASSUMED

Chemical composition
Internal mixture

n(λ), k(λ)



Size.
Shape
Vertical
distribut

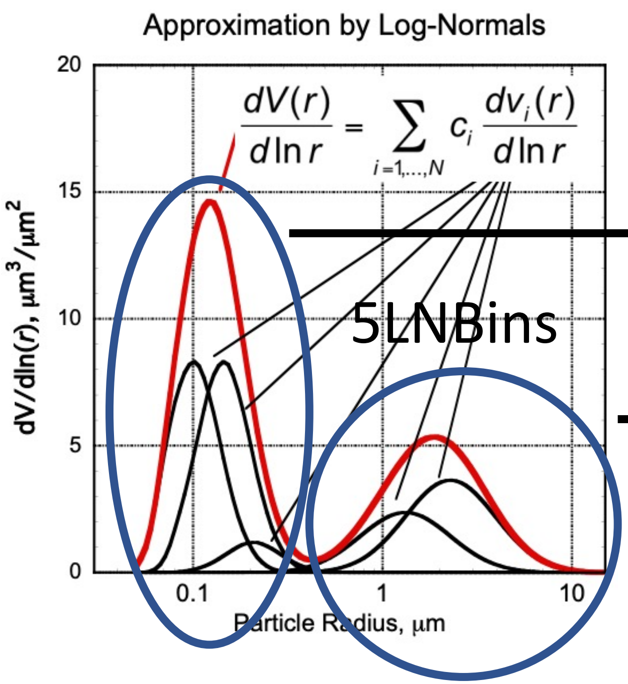


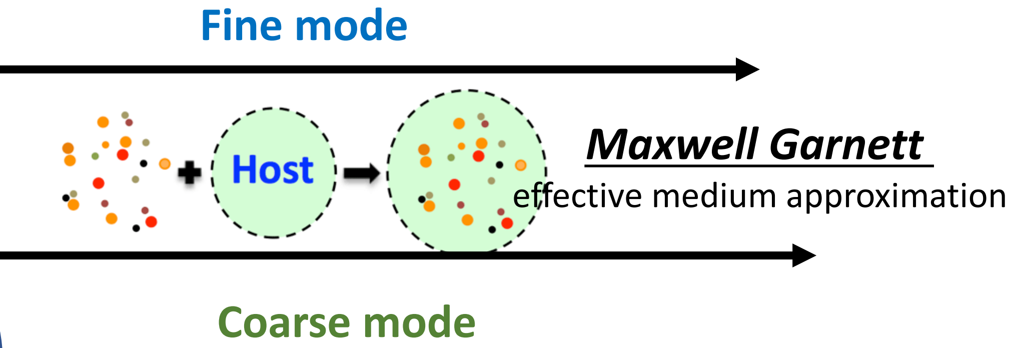
Measurements:

$$\tau_{scat/ext} = \sum_{k=1}^{K}\left(\int_{h_{min}}^{h_{max}}\int_{\varepsilon_{min}}^{\varepsilon_{max}}\int_{r_{min}}^{r_{max}} \frac{C_{scat/ext}\left(n_k(\lambda);k_k(\lambda);h;\varepsilon;r\right)}{V(r)}\frac{dV_k(h)}{dh}\frac{dn_k(r)}{d\varepsilon}\frac{dV_k(r)}{dr}\,dh\,d\varepsilon\,dr\right)$$

Evolution: GRASP Component approach

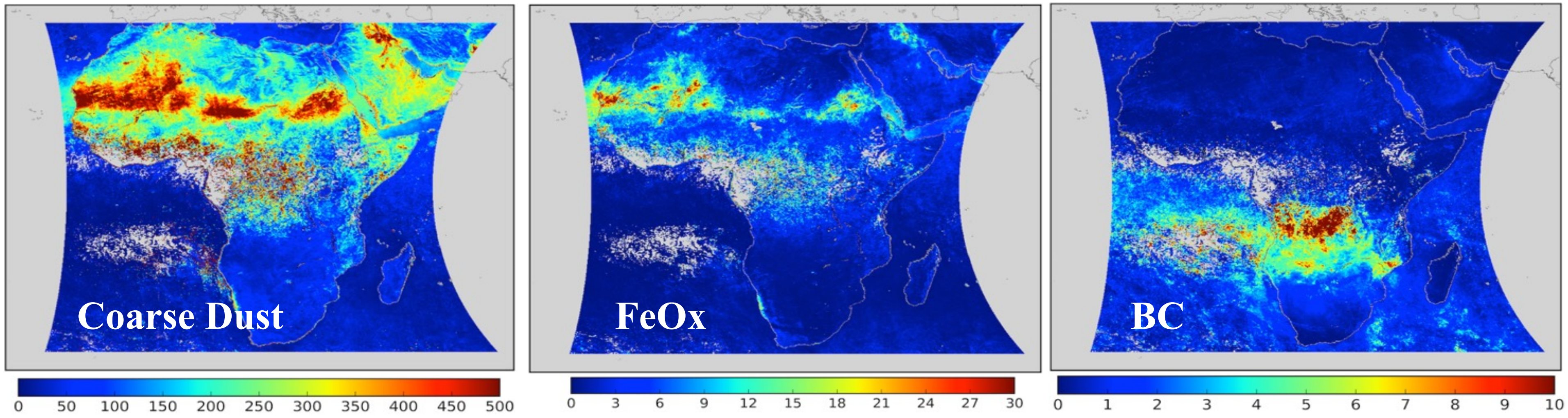
(L. Li et al., ACP, 2019)





BC
BrC
Non-absorbing soluble
Non-absorbing insoluble
Water
Fine mode

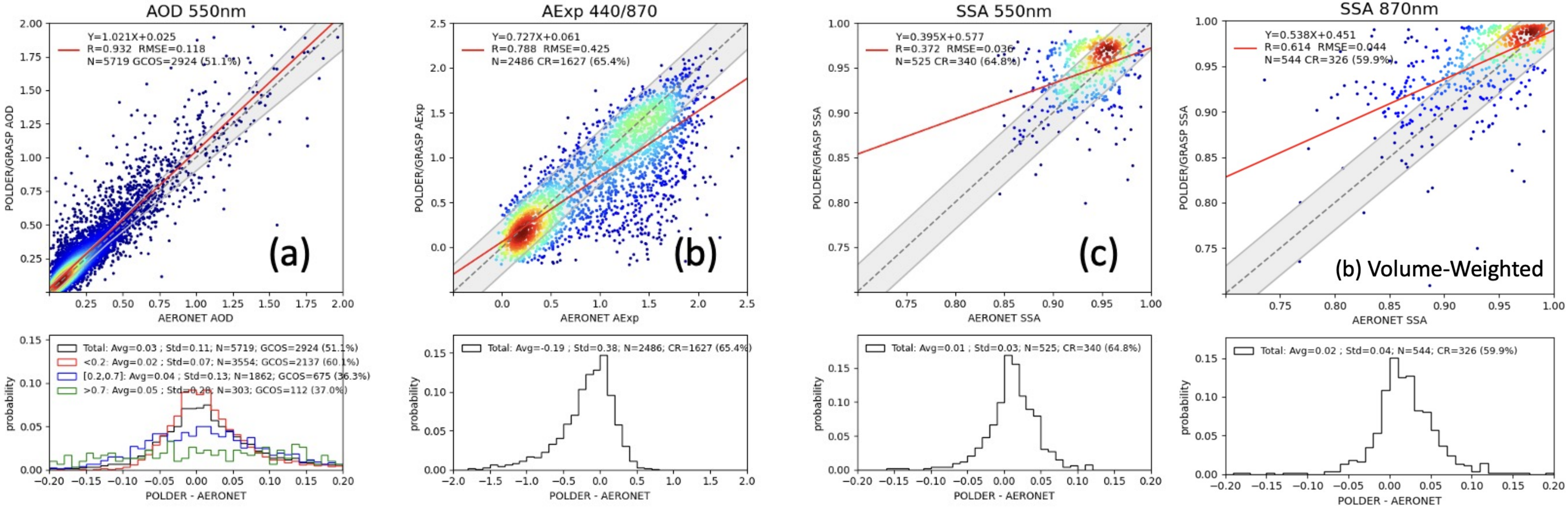
Absorbing insoluble (FeOx)
Non-absorbing insoluble (Dust)
Non-absorbing soluble (SS, etc.)
Water
Coarse mode



Concentrations volumiques (mg/m²) **Automne 2008**

By using ***prescribed spectral refractive index*** of components, *GRASP/Component approach provides consistent and stable results for AOD as well as detailed properties*.

Baseline GRASP **2 modes** approach PARASOL retrievals

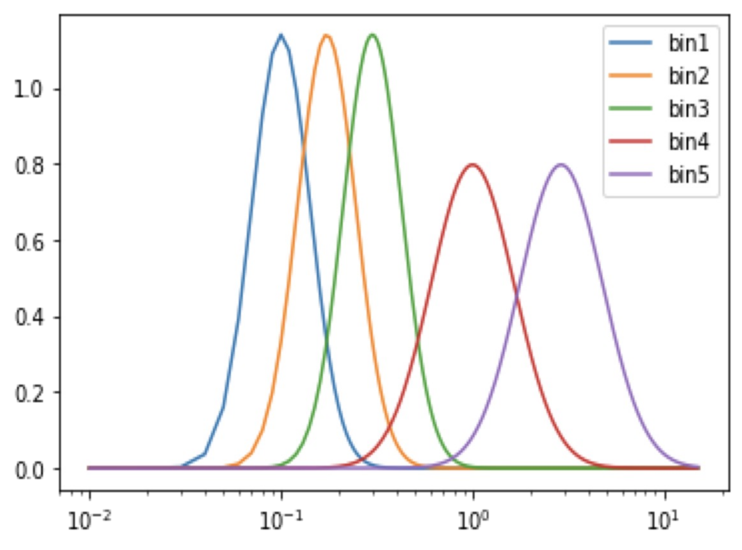


Baseline GRASP aerosol model with 2 modes for 3MI, MAP/CO2M and other sensors

2. GRASP 5LN Chemical Component approach

(L. Li et al., ACP, 2019)

1. 5 LNBins
2. Non-spherisity
3. Chemical Components mixture



	Size distributio n	Volume Concen tration	BC Hydro -philic	BrC Hydro-philic	SU	Wate r	Sea Salt	Dust	
								Iron Oxid e	Quarts
Fine mode	Bin1, Bin2, Bin3	✓	✓	✓	✓	✓			
Coars e mode	Bin4, Bin5	✓				✓	✓	✓	✓

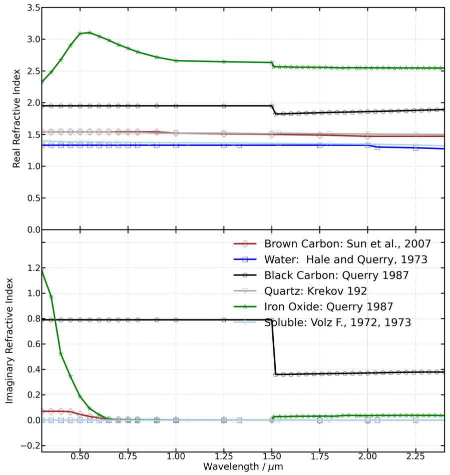
Effective refractive index:

Fine mode :

$$\widehat{m}_{eff}^{fine} = \widehat{m}_{BC}\delta_{BC}^{phil} + \widehat{m}_{BrC}\delta_{BrC} + \widehat{m}_{SU}\delta_{SU} + \widehat{m}_{Water}\delta_{Water}^{fine}$$

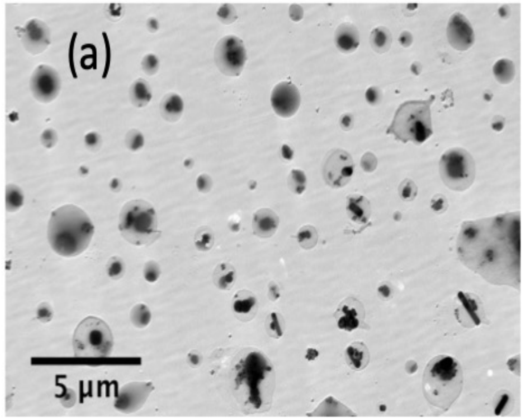
Coarse mode:

$$\widehat{m}_{eff}^{coarse} = \widehat{m}_{Quartz}\delta_{Quartz}^{coarse} + \widehat{m}_{Iron}\delta_{FeOx} + \widehat{m}_{SeaS}\delta_{SeaS} + \widehat{m}_{Water}\delta_{Water}^{coarse}$$

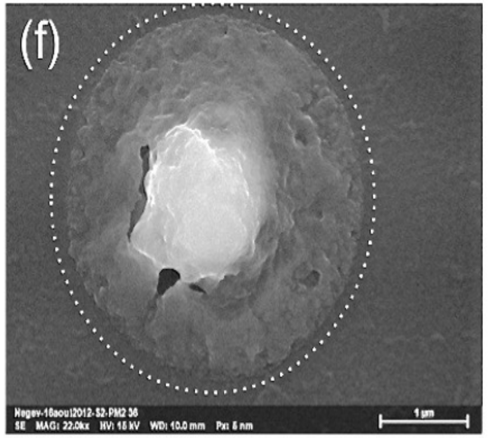


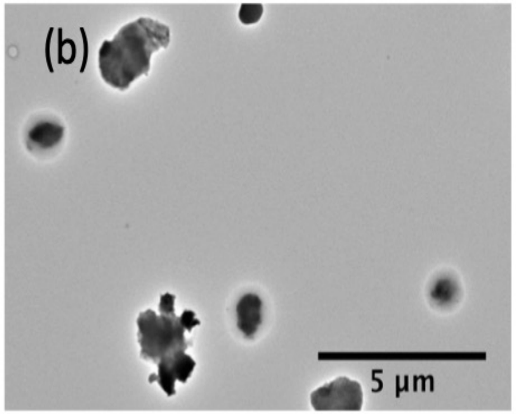
Aerosol non-sphericity and inhomogeneity in remote sensing

Urban aerosol (Lille, France)

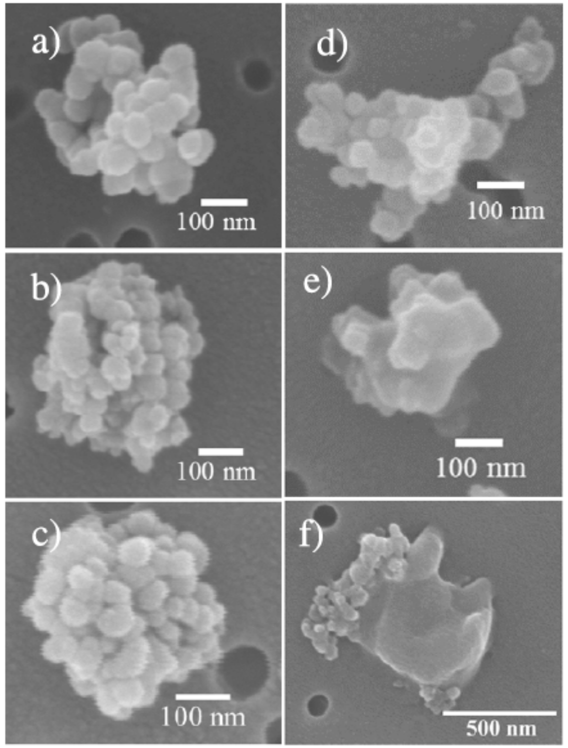


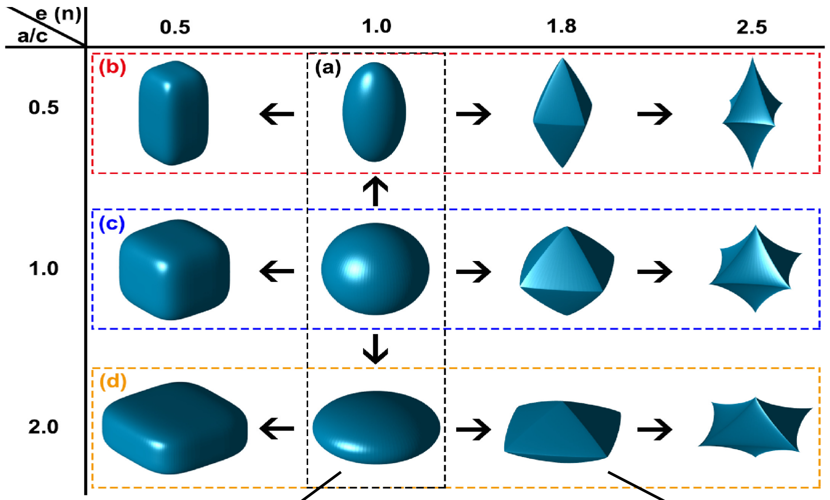
Desert aerosol (Senegal and Israel)





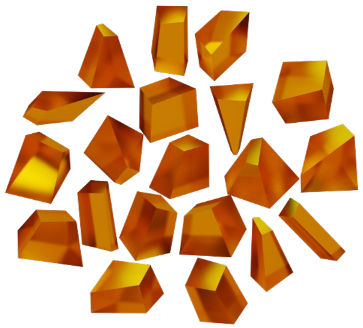
Coated soot





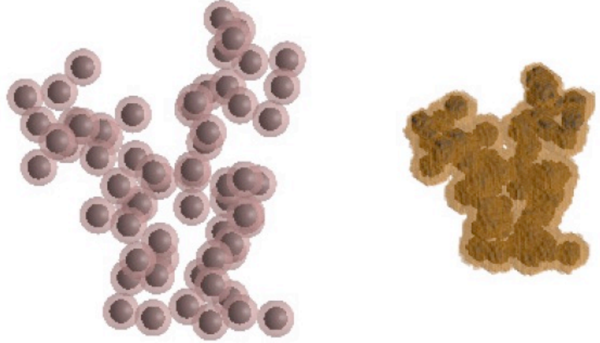
Spheroids
Dubovik et al., 2006

Super Spheroids
Bi et al., 2018



Hexahedrons
Saito et al., 2021 a,b

Model of fractal-like cluster of particles in the shell (under development in collaboration with V. Tyshkovets and L. Berdina)

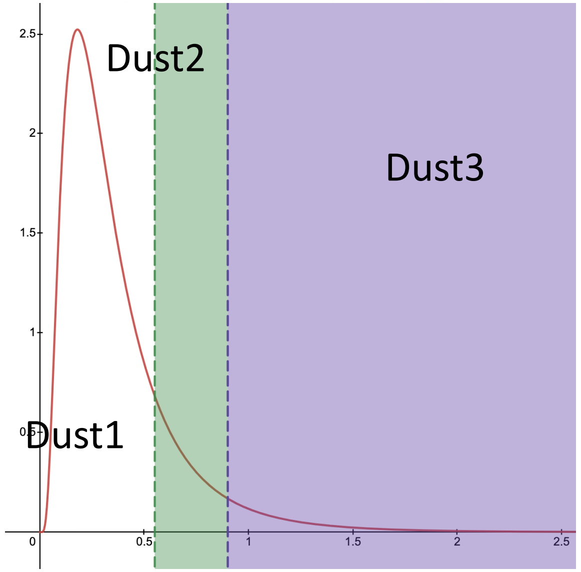


Size distribution in CAMS/MERRA-2 versus GRASP

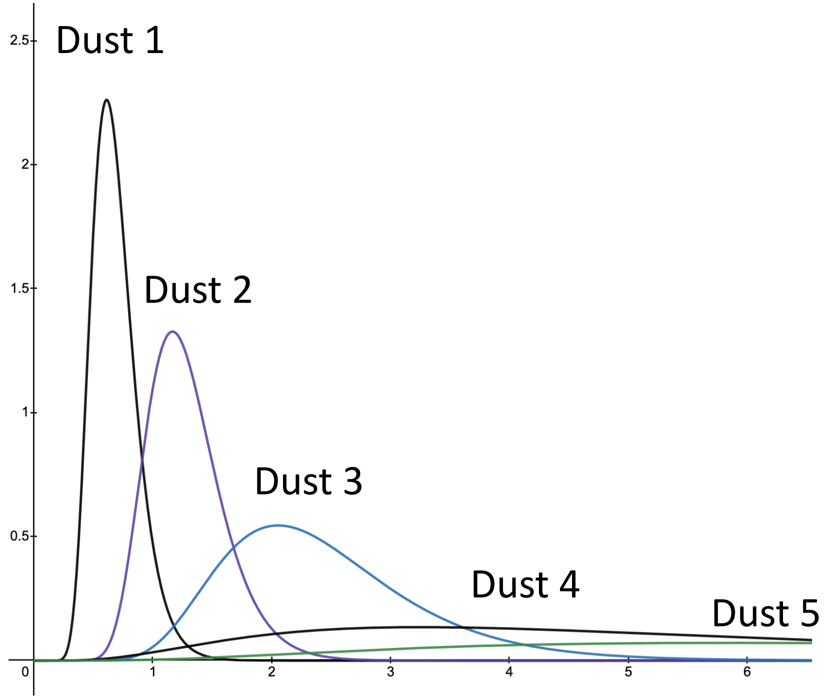
- The way of cutting the size distribution to define bins for Dust and Sea Salt in CAMS looks very different from the one used in remote sensing retrieval where SD is smooth function.

Dust bins from CAMS (OC and up to cycle 47R3)

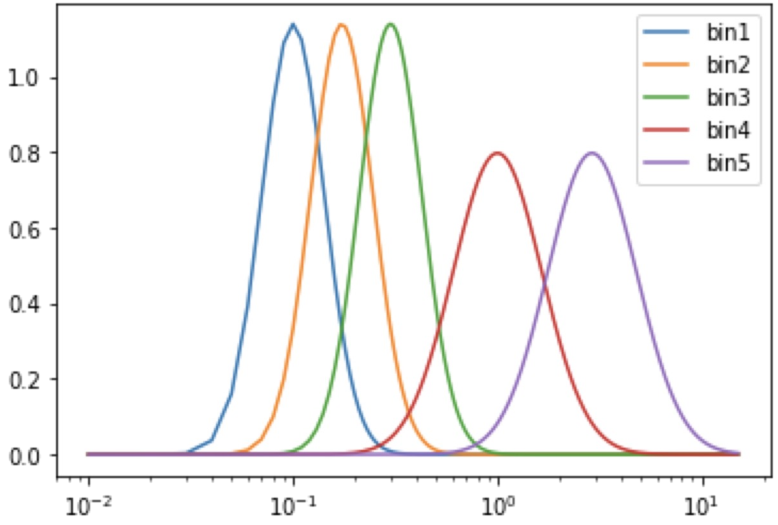
$$n(r) = \frac{\mathrm{d}N(r)}{\mathrm{d}r} = \frac{N}{\sqrt{2\pi}\, r \ln(\sigma)} \exp\left(-\frac{\ln^2(r/r_{\mathrm{mod}})}{2\ln^2(\sigma)}\right)$$



Dust tracer from MERRA2 Natural Run



GRASP 5 LN SD bins



Harmonization of remote sensing aerosol with aerosol in CAMS/MERRA-2:

GRASP / remote sensing

2 modes

Optical properties
(*refractive indices*)

≠

CAMS / MERRA-2

5 modes

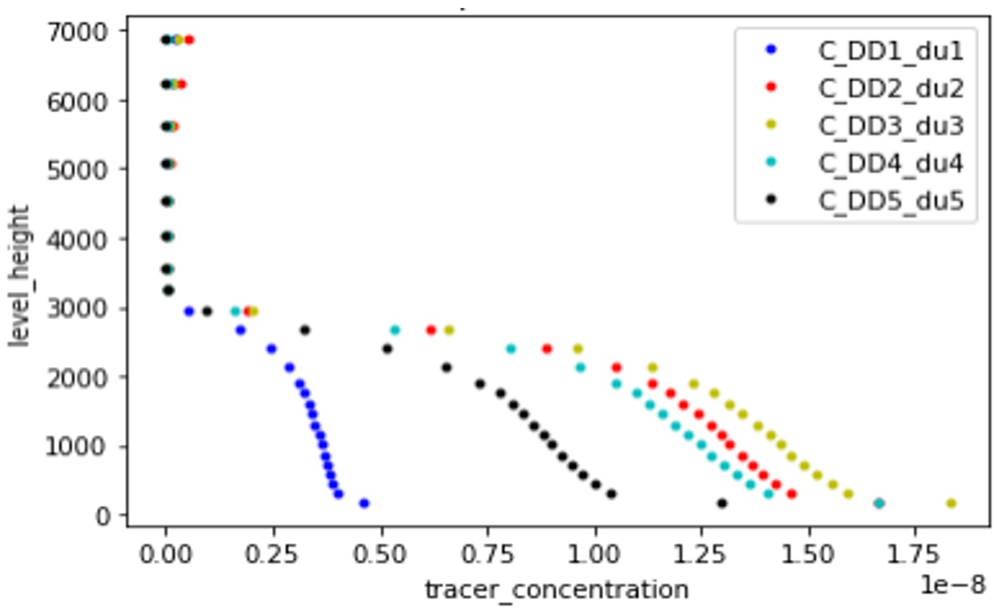
Optical properties
(*refractive indices*)

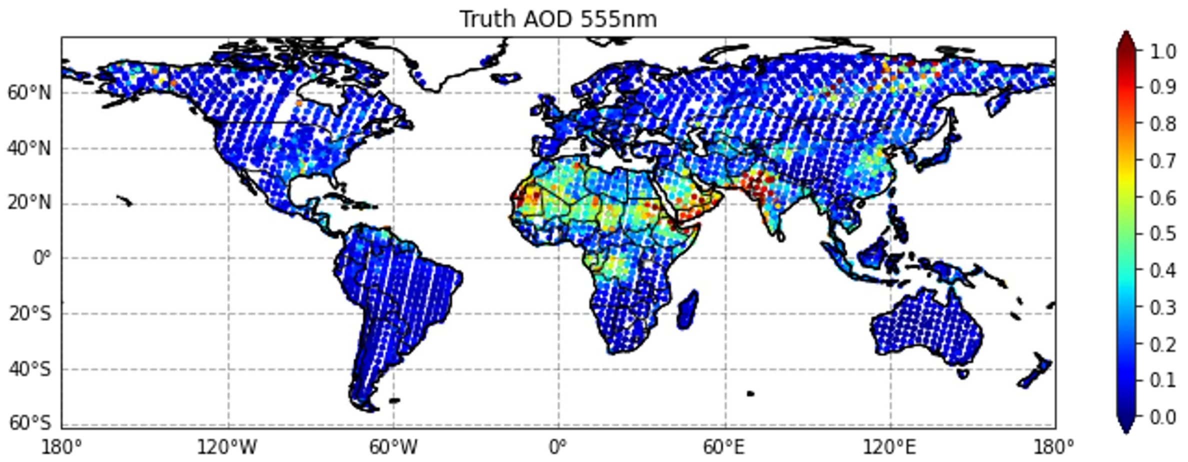
Feasibility tests	Harmonization toward CAMS aerosol representation
1	SU in separate mode
2	Hydrophobic BC and BrC in the separate modes
3	Sea Salt and Dust in separates modes
4	Adjustment of the complex refractive index of GRASP aerosol chemical components

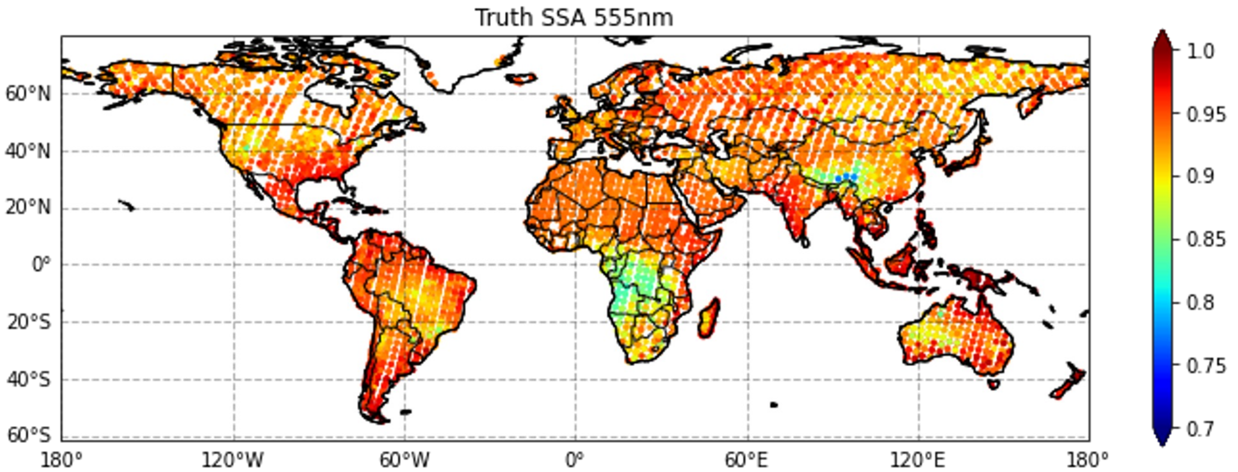
Synthetic dataset

Based on CAMS aerosol model

- Pressure, Relative, humidity profiles
- Mass mixing ratio
- 11 aerosol tracers and level concentration for 5 aerosol species : Sulphate (SU), Desert dust (DU), Sea Salt (SS), Organic (OC) and Black Carbon (BC)

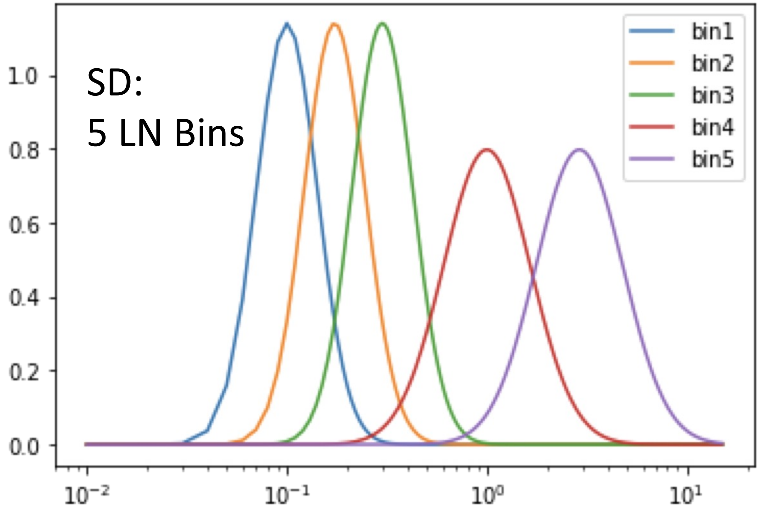






1.1 **3 aerosol modes**: Hydrophilic and Hydrophobic **BC and BrC** in separate modes

	Size distribution	Volume Concentration	Volume fraction of chemical components								
			BC Hydro-phobic	BrC Hydro-phobic	BC Hydro-philic	BrC Hydro-philic	SU	Water	Dust		Sea Salt
									Iron Oxide	Quarts	
Fine mode 1	Bin1, Bin2	✓	✓	✓							
Fine mode 2	Bin2, Bin3	✓			✓	✓	✓	✓			
Coarse mode	Bin4, Bin5	✓						✓	✓	✓	✓



Effective refractive index:

Fine mode 1:

$$\hat{m}_{eff}^{fine_1} = \hat{m}_{BC}\delta_{BC}^{phob} + \hat{m}_{BrC}\delta_{BrC}^{phob}$$

Fine mode 2:

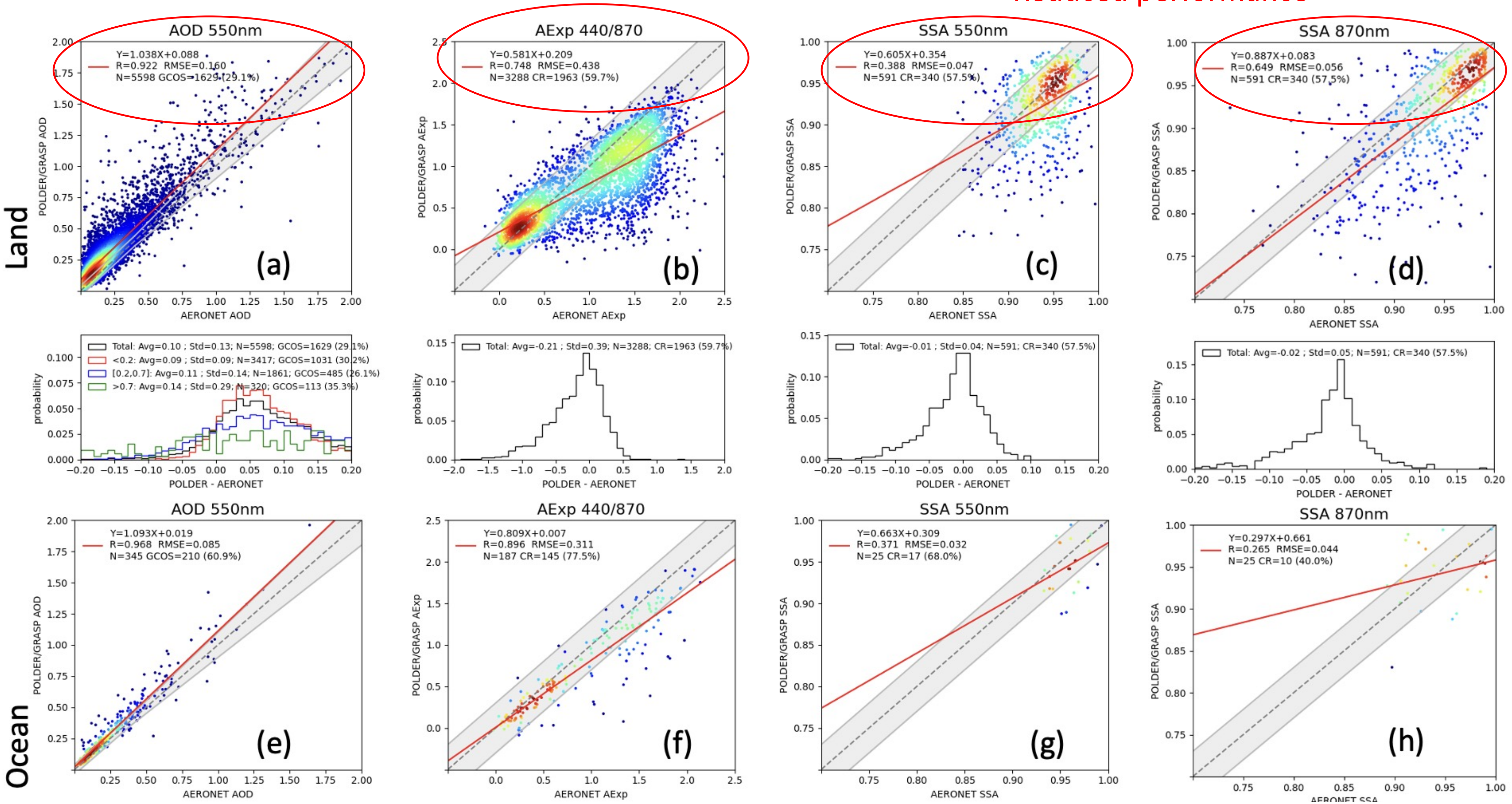
$$\hat{m}_{eff}^{fine_2} = \hat{m}_{BC}\delta_{BC}^{phil} + \hat{m}_{BrC}\delta_{BrC}^{phil} + \hat{m}_{SU}\delta_{SU} + \hat{m}_{Water}\delta_{Water}^{fine}$$

Coarse mode:

$$\hat{m}_{eff}^{coarse} = \hat{m}_{Quartz}\delta_{Quartz}^{coarse} + \hat{m}_{Iron}\delta_{FeOx} + \hat{m}_{SeaS}\delta_{SeaS} + \hat{m}_{Water}\delta_{Water}^{coarse}$$

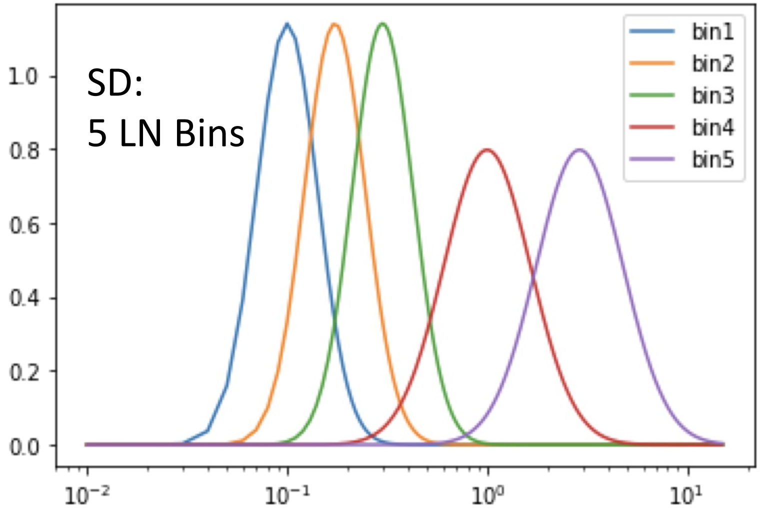
3 aerosol modes: Hydrophilic and Hydrophobic **BC and BrC** in separate modes (performance on PARASOL measurements)

Reduced performance



1.2 3 aerosol modes: Dust and Sea Salt in separate modes

	Size distribution	Volume Concentration							
			BC Hydro-philic	BrC Hydro-philic	SU	Water	Sea Salt	Dust	
								Iron Oxide	Quarts
Fine mode	Bin1, Bin2	✓	✓	✓	✓	✓			
Coarse mode 1	Bin2, Bin3	✓				✓	✓		
Coarse mode 2	Bin4, Bin5	✓						✓	✓



Effective refractive index:

Fine mode:

$$\hat{m}_{eff}^{fine} = \hat{m}_{BC}\delta_{BC}^{phil} + \hat{m}_{BrC}\delta_{BrC} + \hat{m}_{SU}\delta_{SU} + \hat{m}_{Water}\delta_{Water}^{fine}$$

Fine mode 2:

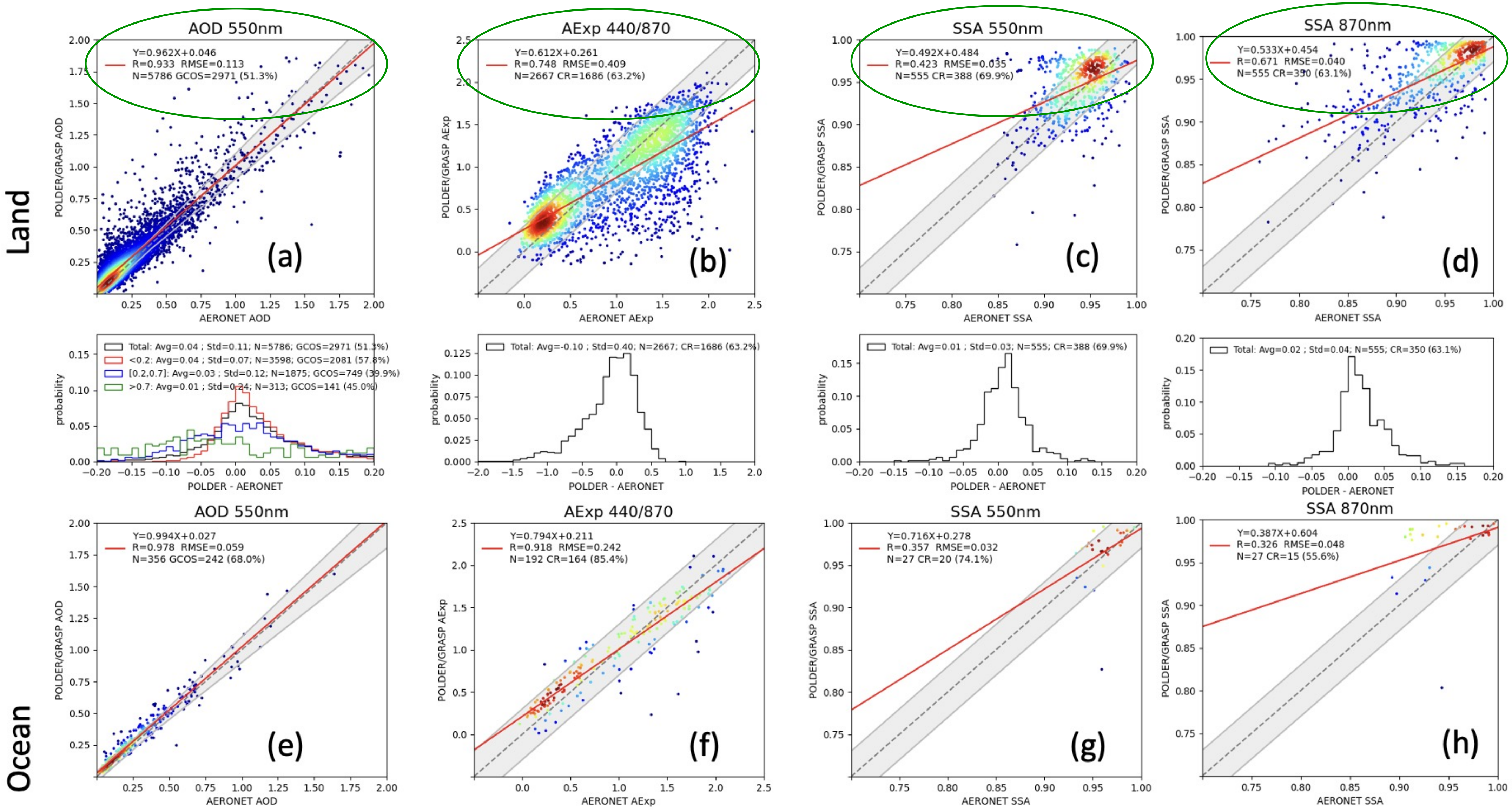
$$\hat{m}_{eff}^{coarse_1} = \hat{m}_{SeaS}\delta_{SeaS} + \hat{m}_{Water}\delta_{Water}^{coarse}$$

Coarse mode:

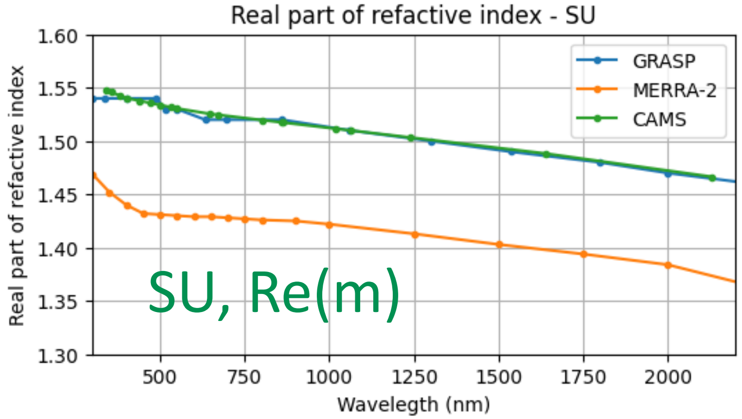
$$\hat{m}_{eff}^{coarse_2} = \hat{m}_{Quartz}\delta_{Quartz}^{coarse} + \hat{m}_{Iron}\delta_{FeOx}$$

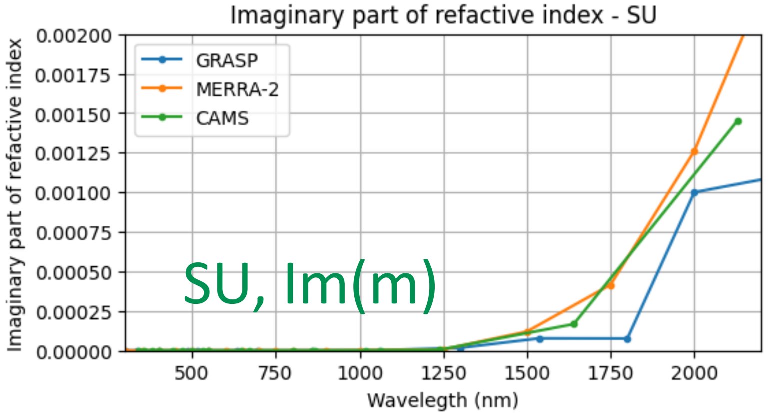
Dust and Sea Salt in different modes: (performance on PARASOL measurements)

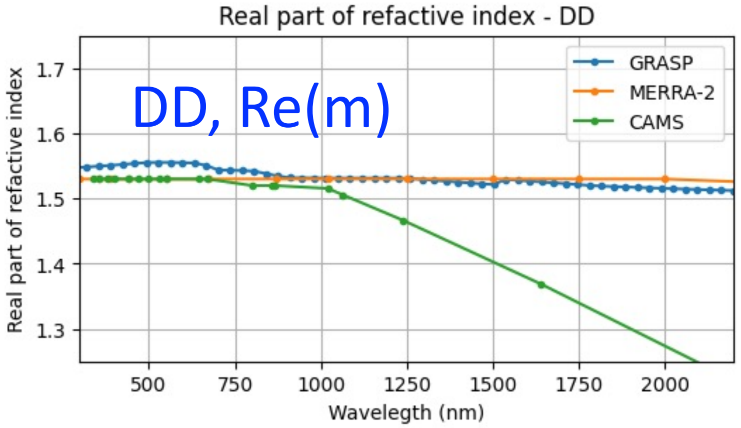
Improved performance

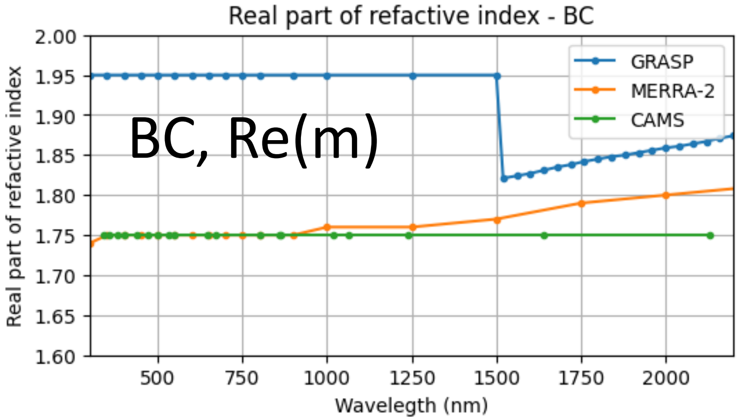


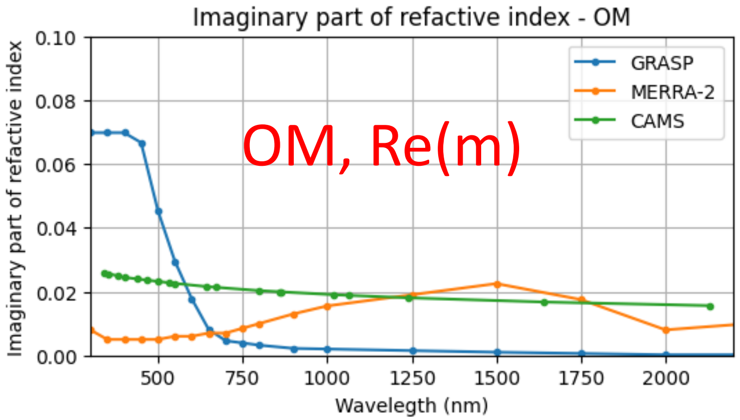
Refractive indices for aerosol species

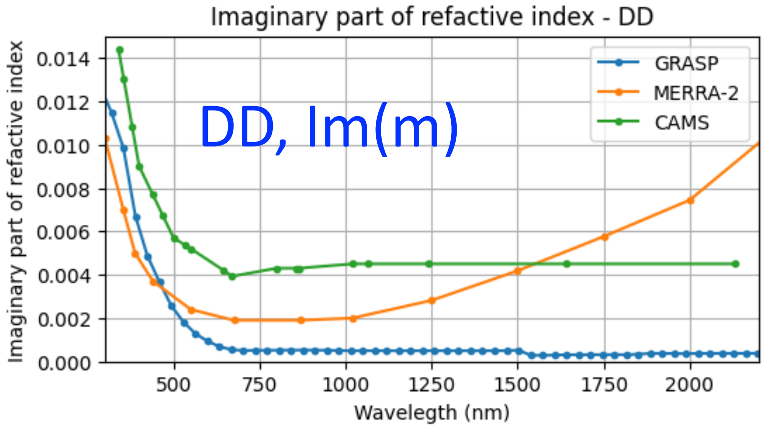


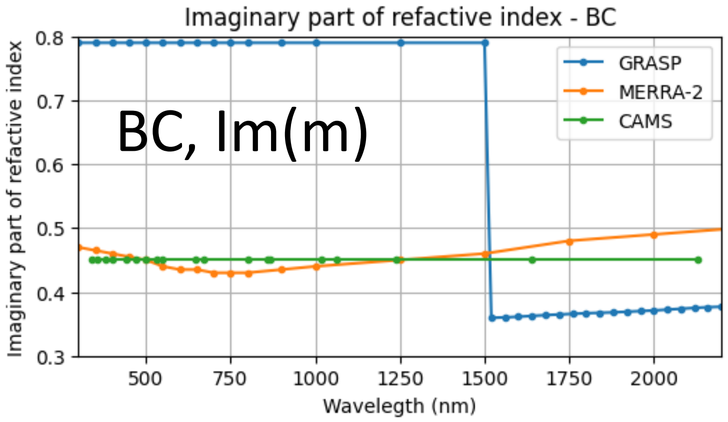


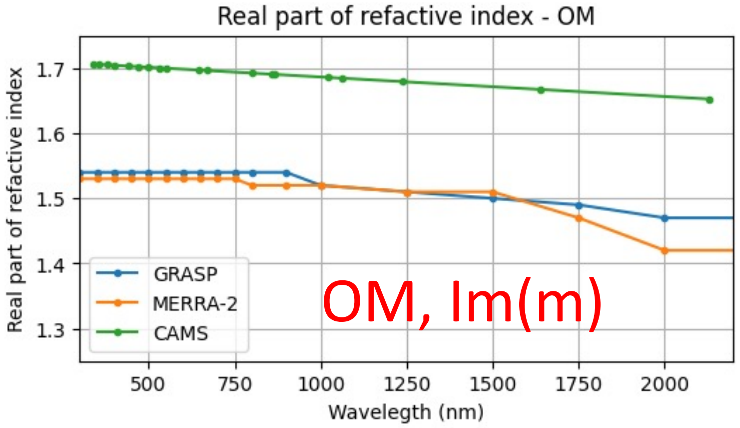






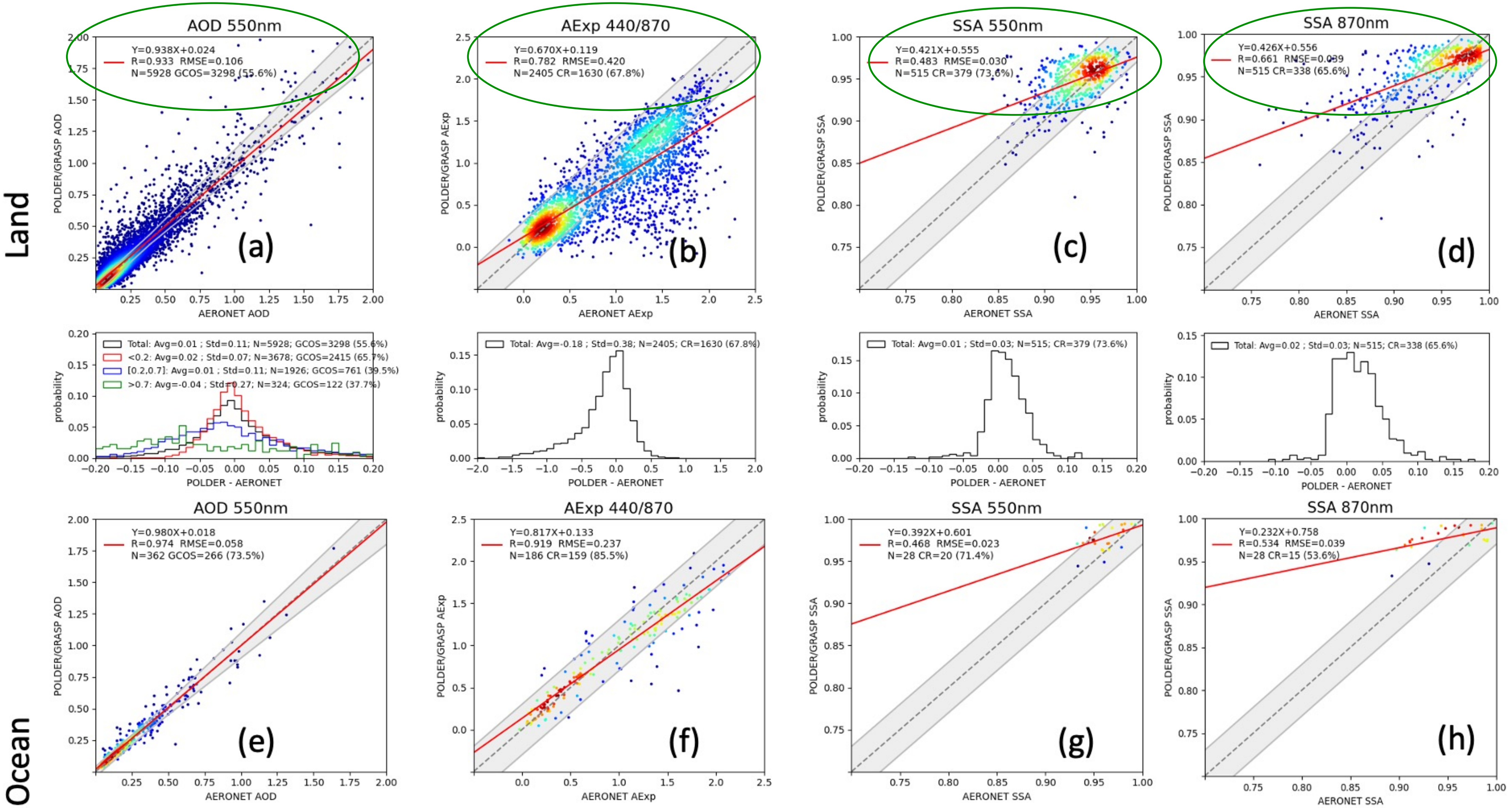




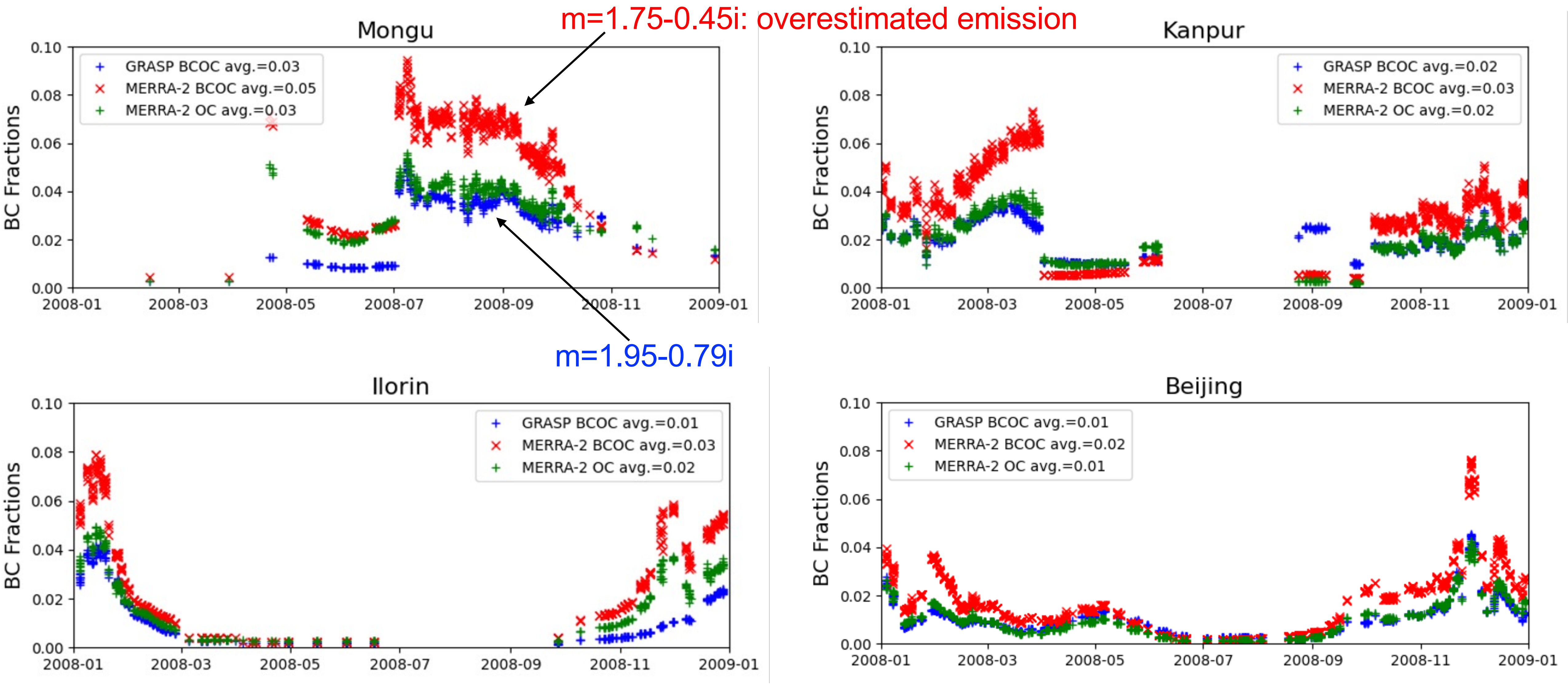


Refractive index harmonization: MERRA-2/CAMS BC and OM refractive index in GRASP

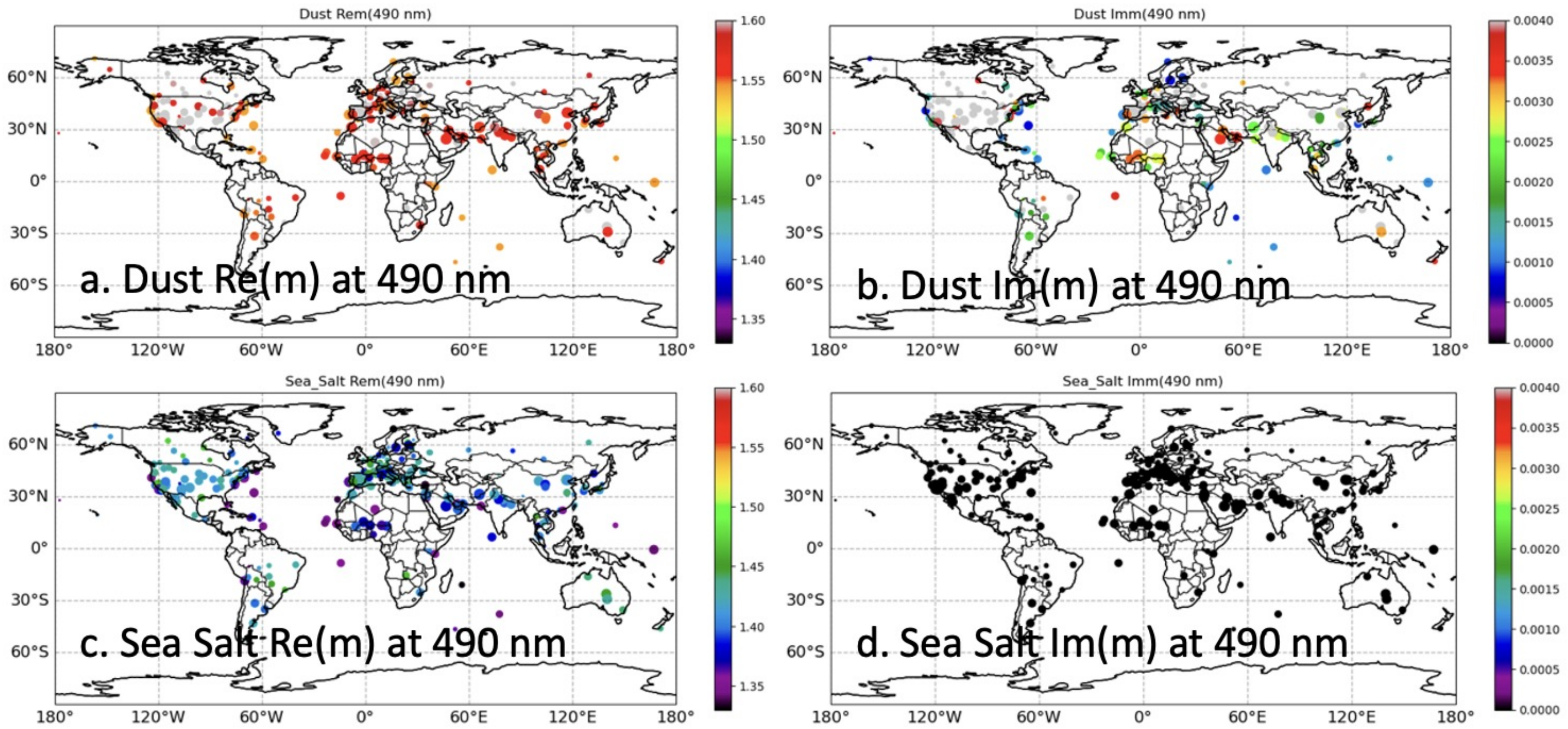
Similar performance

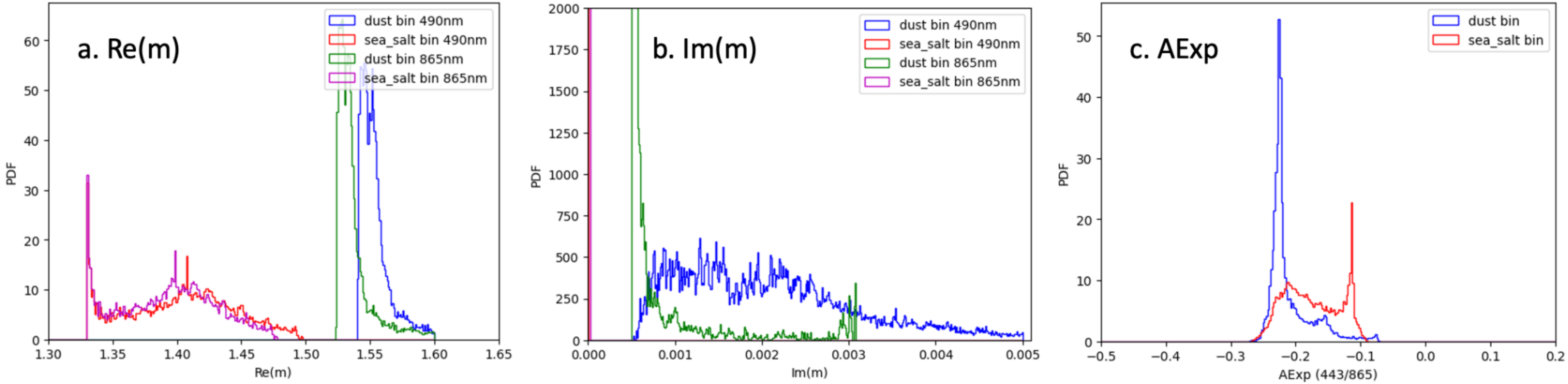


Refractive index harmonization: MERRA-2/CAMS BC and OM refractive index in GRASP



Dust and Sea Salt in different modes: statistic over AERONET stations





Summary on complex refractive index studies

Harmonization of the complex refractive index in GRASP and CAMS and MERRA-2 models showed:

Feasibility tests with real PARASOL data	Performance in **AOD**	Performance in **AE**	Performanc e in **SSA**
Adjustment of the complex refractive index of aerosol	Same quality	Same quality	Same quality

REDUNDENCY

- Great agreement in synthetic retrieval;
- NO EFFECT on optical properties retrieved from real PARASOL data;
- Overestimation of emission derived from retrieved optical properties retrieved from real PARASOL;

Current status of harmonization:

- Harmonization of remote sensing and climate models is relatively straightforward.
- Rather clear relation between parameters can be set up.

Current Questions:

CAMS / MERRA-2:

- assumptions of species ref. indices - ?;
- variability of DD optical properties - ?;
(no variability in ref. index)
- non-sphericity of DD -?

- inhomogeneity of aerosol -?
(externa vs internal, optics vs transport physics);

GRASP / remote sensing

- is 3 external mode optimal - ?;
(there are redundant parameter for optics)
- which parameters should be in focus:
(AOD (λ), AE, SSA(λ), AODF ?, else -?)
- which properties can be adapted from CAMS
(aerosol profiles, profiles of relative humidity, etc.?)

No degradation of real retrieval from modification!

Aerosol models in CAMS/MERRA-2 and remote sensing

	Aerosol modelling approach	CAMS/MERRA-2	Optimal model for <u>multi-angular polarimetric</u> remote sensing	Single/bi-viewing imagers	Lidars
1	**Aerosol modes and aerosol species**	• **5 aerosol species**, **7 components**: BC(2), OC(2), SU, SS(3-5), Dust (3-5) • External mixture	• **5-7 aerosol species** distributed in **2-3 different aerosol modes** • External, Internal or hybrid mixture	• ? • ? • ?	• ? • ? • ?
2	**Refractive index**	• Fixed for each dry specie	• Fixed for each component • Can be retrieved from internal mixture or at each wavelength	• ? • ? • ?	• ? • ? • ?
3	**Aerosol vertical profile**	• Mass Mixing ratio for each tracer (bin) at each level • Vertical dependence of aerosol characteristics with RH	1-3 concentration profiles 1: the same for all modes 2: different for fine and coarse modes 3: different for each aerosol mode	• ? • ? • ?	• ? • ? • ?
4	**Size distribution/** Hygroscopicity	• SD for each aerosol bin • SD parameters change with RH	• Size distribution parameters • A few bins for each of 1-3 modes	• ? • ? • ?	• ? • ? • ?
5	**Non-sphericity/ inhomogeneity**	Not accounted yet	May be accounted using different models	?	• ?

THANK YOU !

Aerosol models in CAMS/MERRA-2 and remote sensing

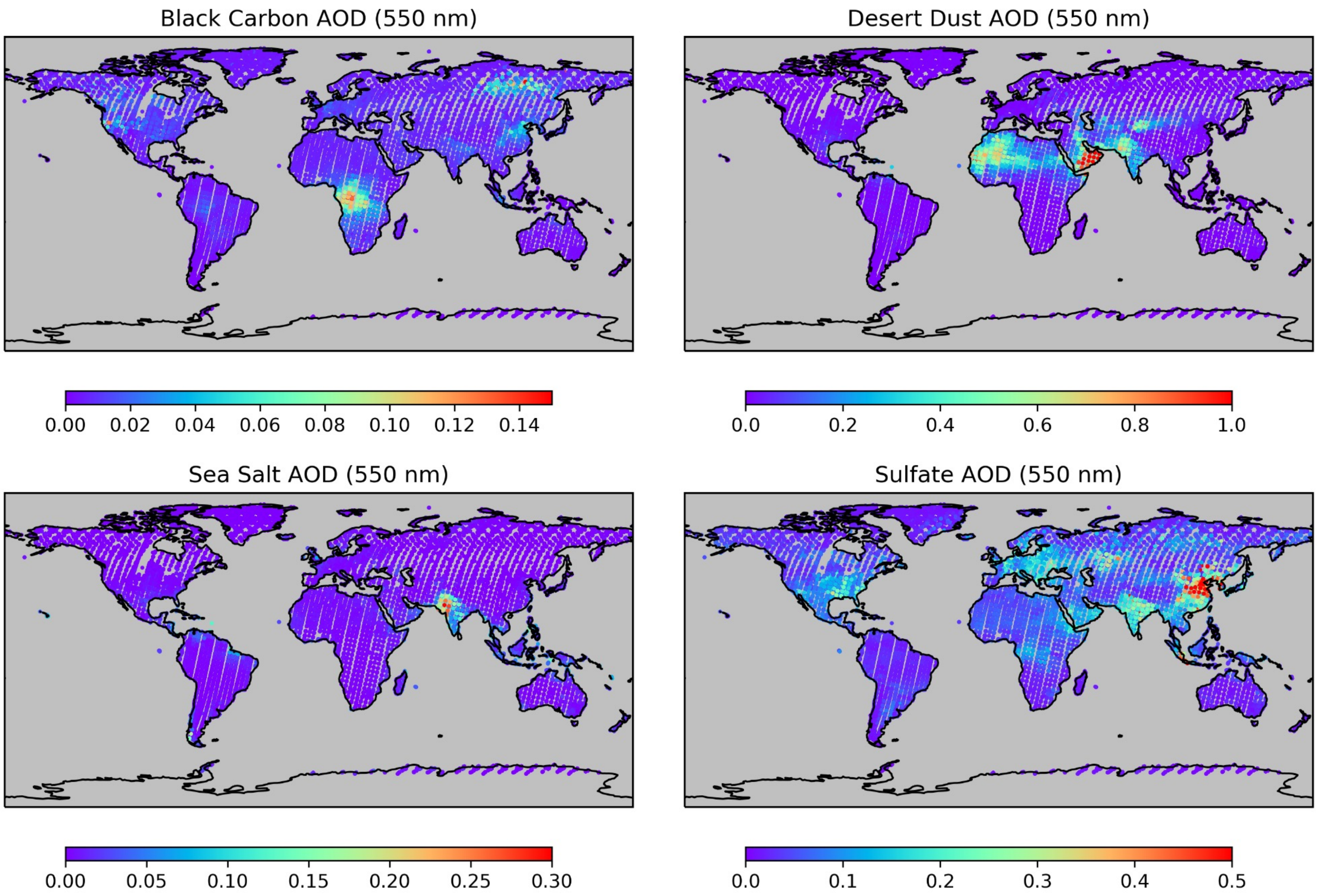
	Aerosol modelling approach	CAMS/MERRA-2	Optimal model for multi-angular polarimetric remote sensing
1	Aerosol modes and aerosol species	• **5 aerosol species, 7 components**: BC(2), OC(2), SU, SS(3-5), Dust (3-5) • External mixture	• **5-7 aerosol species** distributed in **2-3 different aerosol modes** • External, Internal or hybrid mixture
2	Refractive index	• Fixed for each dry specie	• Fixed for each component • Can be retrieved from internal mixture or at each wavelength
3	Aerosol vertical profile	• Mass Mixing ratio for each tracer (bin) at each level • Vertical dependence of aerosol characteristics with RH	1-3 concentration profiles 1: the same for all modes 2: different for fine and coarse modes 3: different for each aerosol mode
4	**Size distribution/** Hygroscopicity	• SD for each aerosol bin • SD parameters change with RH	• Size distribution parameters • A few bins for each of 1-3 modes
5	**Non-sphericity/ inhomogeneity**	Not accounted yet	May be accounted in different models

Harmonization questions to answer

	Aerosol modelling approach	CAMS/MERRA-2	Multi-angular polarimetric remote sensing
1	**Aerosol modes and aerosol species**	➢ How complete representation of aerosol with 5 main species: BC, OC, SU, SeaSalt, Dust? ➢ External mixture ➢ What aerosol characteristics are most crucial to be accounted for harmonization: • total AOD or AOD for each species? • Fine mode AOD? • Angstrom Exponent? • SSA? • What spectral bands?	➢ Redundancy of external mixture if internal one for the same components is presented: • Can sensitivity to different aerosol species be increased with spectral measurements in UV and SWIR, TIR (AERONET, 3MI, IASI)?
2	**Refractive index**	➢ How it is representative for each aerosol specie?	➢ More physical dependence of the effective refractive index on the Relative Humidity or water vapour content.
3	**Aerosol vertical profile**	➢ How reliable mass mixing ratio in CAMS and can it be used as a priori estimates in the remote sensing retrieval?	➢ Accounting for aerosol microphysics vertical dependence, for example, for the synergy of polarimetric and LIDAR measurements. ➢ How detailed vertical aerosol microphysics profile is important for passive instruments?
4	**Size distribution/** Hygroscopicity	➢ How representative SD for one mode SD species: BC, OC, SU in CAMS?	➢ Physically based accounting for aerosol hygroscopic growth with relative humidity.
5	**Non-sphericity/ inhomogeneity**	➢ How important non-sphericity/inhomogeneity in CAMS and how it may affect the atmospheric radiance/flux calculations?	➢ Accounting for particle inhomogeneity

Harmonization questions to answer:

1. Main aerosol component: BC, OM, SU, Sea Salt, Dust.
   - How complete this representation of aerosol?
   - External, internal or hybrid mixture?
   - Optimal balance between complexity of aerosol preorientation and number of retrieved parameters in remote sensing.
2. The spectral dependence for each component.
   - How representative it is?
3. Vertical profile
   - How reliable mass mixing ratio in CAMS and can it be used as a priori estimates in the retrieval?
   - The effect of vertical dependence of aerosol characteristics vs column averaged properties.
4. SD for aerosol
   - How representative and flexible it is in CAMS?
   - Retrieved SD in remote sensing vs prescribed with accounting for hygroscopic growth in CAMS
5. Non-sphericity and inhomogeneity in CAMS
   - How important it is in CAMS and how it may affect the atmospheric radiance calculations
6. What are the main aerosol parameters for harmonization with CAMS?
   - AOD? What spectral bands? Fine mode AOD? Angstrom Exponent? SSA?


Black Carbon AOD (550 nm)
0.00 0.02 0.04 0.06 0.08 0.10 0.12 0.14
Desert Dust AOD (550 nm)
0.0 0.2 0.4 0.6 0.8 1.0
Sea Salt AOD (550 nm)
0.00 0.05 0.10 0.15 0.20 0.25 0.30
Sulfate AOD (550 nm)
0.0 0.1 0.2 0.3 0.4 0.5

CAMS aerosol model in IFS

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https://doi.org/10.5194/gmd-13-1007-2020

Geoscientific Model Development
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An aerosol climatology for global models based on the tropospheric aerosol scheme in the Integrated Forecasting System of ECMWF

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Geoscientific Model Development
Open Access

Description and evaluation of the tropospheric aerosol scheme in the Integrated Forecasting System (IFS-AER, cycle 47R1) of ECMWF

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Copernicus Atmosphere Monitoring Service

Review of optical properties and size distribution

Deliverable report for D1.4.3

Issued by: HYGEOS/Samuel Remy

Date: 23/07/2021

Ref: CAMS43_2021SC2_D1.4.3_202106_optical_v3

The most optimal GRASP aerosol approach based on retrieval of PARASOL measurements

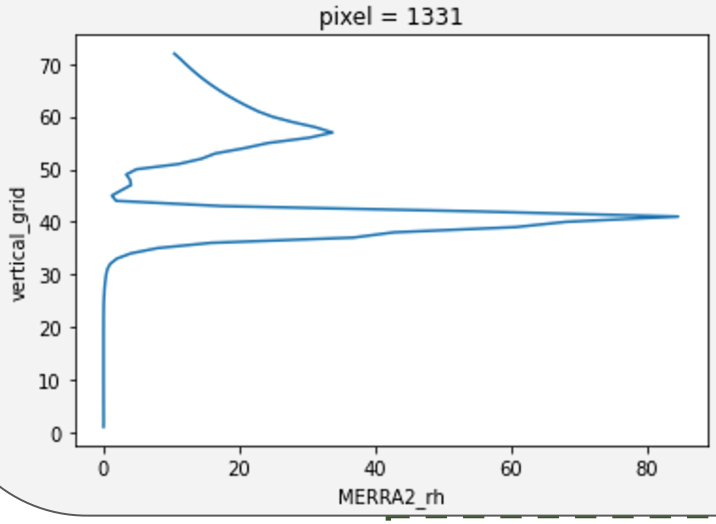
		GRASP chemical components with ***external*** mixture of 3 modes (~10-15 parameters)							
		Mode 1 (*internal mixture*, ***3 SD bins***)				Mode 2 (*internal mixture*, ***2 SD bins***)		Mode 3 (*internal mixture*, ***2 SD bins***)	
		BC δ_{BC}	BrC δ_{BrC}	Sulfate	Water δ^{fine}_{Water}	Water δ^{coarse}_{Water}	Sea salt	FeOx δ_{Iron}	Quartz δ^{coarse}_{Quartz}
***External* mixture** of Aerosol Species in CAMS or MERRA-2 (11-15 parameters)	BC Hydrophobic								
	BC Hydrophilic								
	OM Hydrophobic								
	OM Hydrophilic	X	X	X	X				
	SU								
	Sea Salt **(3 bins)**					X	X		
	Dust **(3 bins)**							X	X

Redundancy of external mixture when internal one for the same components is taken into account?

3.1 Vertical profiling. Transport models

For each pixel:

- 72 vertical level height
- Pressure profiles
- Relative humidity
- Mass mixing ratio
- 15 aerosol tracer level concentration



CAMS/MERRA-2 aerosol components k for RH set $\{RH_j\}$ and wavelength set $\{\lambda_i\}$:

1. Phase matrix: $P_k(RH_j, \lambda_i)$
2. Mass extinction coefficient: $\beta_{ext}^{k}(RH_j, \lambda_i)$
3. SSA: $\omega_{ext}^{k}(RH_j, \lambda_i)$ (or $\beta_{sc}^{k}(RH_j, \lambda_i)$)

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3. SSA: $\omega_{ext}^{k}(RH_j, \lambda_i)$ (or $\beta_{sc}^{k}(RH_j, \lambda_i)$)

3.2 Vertical profiling. Remote sensing retrieval

1. Column scattering and extinction cross sections:

$$< C_{v\,sc}^{(k)} >= \frac{\int_{z_1}^{z_{l max}} C_{v\,sc}^{(k)}(z')c_v^{(k)}(z')\,dz'}{\int_{z_1}^{z_{l max}} c_v^{(k)}(z')\,dz'} \qquad < C_{v\,ext}^{(k)} >= \frac{\int_{z_1}^{z_{l max}} C_{v\,ext}^{(k)}(z')c_v^{(k)}(z')\,dz'}{\int_{z_1}^{z_{l max}} c_v^{(k)}(z')\,dz'}$$

2. Column averaged Phase matrix:

$$< \mathbf{P}_k >= \frac{\int_{z_1}^{z_{l max}} \mathbf{P}_k C_{v\,sc}^{(k)}(z')c_v^{(k)}(z')\,dz'}{\int_{z_1}^{z_{l max}} C_{v\,sc}^{(k)}(z')c_v^{(k)}(z')\,dz'}$$

3. Recalculated extinction coefficient corresponds to the case RT calculations are performed for vertically averaged aerosol single scattering properties:

$$< \alpha_l^{(k)}\ (z) > =< C_{v\,ext}^{(k)} > c_v^{(k)}(z)$$

3. Vertical profile of the concentration:

$$c_v^{(k)}(z) = c_{0v}^{(k)} \frac{1}{h} exp\left(-\frac{z}{h}\right)$$

3.3 CAMS/MERRA-2 MMR and volume concentration for each aerosol mode: easy to setup the relations

AOD for each tracer (bin) *k*:

$$\Delta\tau_k^* = \int_{z_1}^{z_{l_{max}}} C_{v\,ext}^{(k)}(z')\, c_v^{(k)}(z')\, dz'$$

$c_v^{(k)}(z')$ is volume concentration retrieved by GRASP

$M_{mix}^{(k)}(z)$ is aerosol mass mixing ratio From CAMS or MERRA-2

$$c_v^{(k)}(z) = \left(M_{mix}^{(k)}(z)\, \rho_{air}(z)\right) \frac{\left[f_{grow}^{(k)}(z)\right]^3}{\rho_{dry}^{(k)}}$$

$f_{grow}^{(k)}(z)$ aerosol size growth factor, $\rho_{air}(z)$ is air density, $\rho_{dry}^{(k)}$ is dry aerosol density