Harmonization of the aerosol modeling in CAMS model and remote sensing approaches

Atmosphere Monitoring

AMEO

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2 - GRASP-SAS, Remote sensing developments, Lezennes, France,
3- NASA/Langley, Hampton, USA 4- EUMETSAT, Darmstadt, Germany
, 5 – ECMWF, Reading, UK

GRASP

Laboratoire d'Optique Atmosphérique

EUMETSAT

ECMWF

NASA

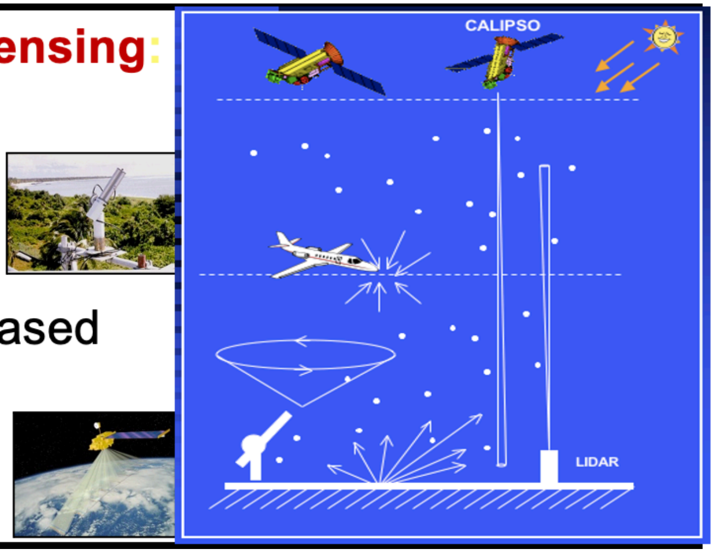
MIRA Webinar, on-line, 22 April 2024

Motivation:

Atmosphere Monitoring

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

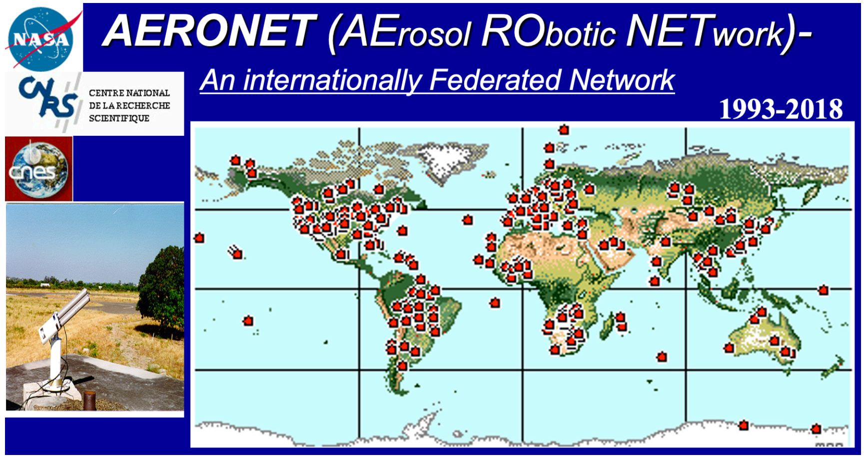
MERRA-2

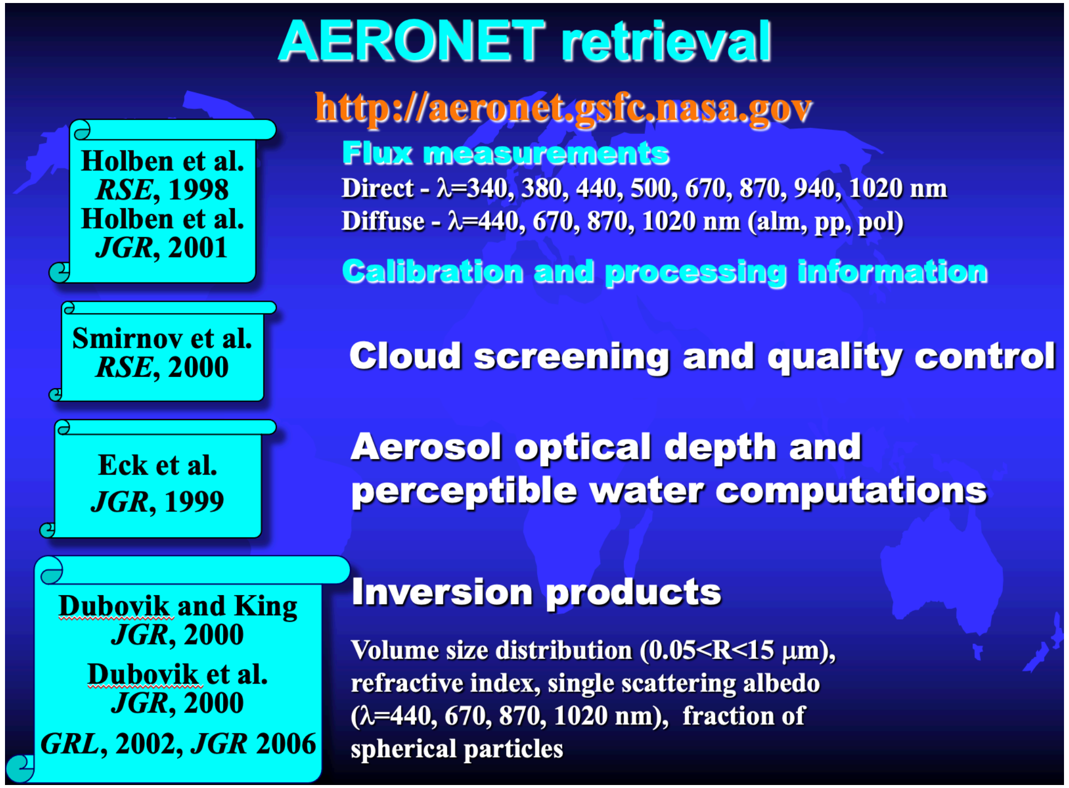
MERRA-2 Natural Run

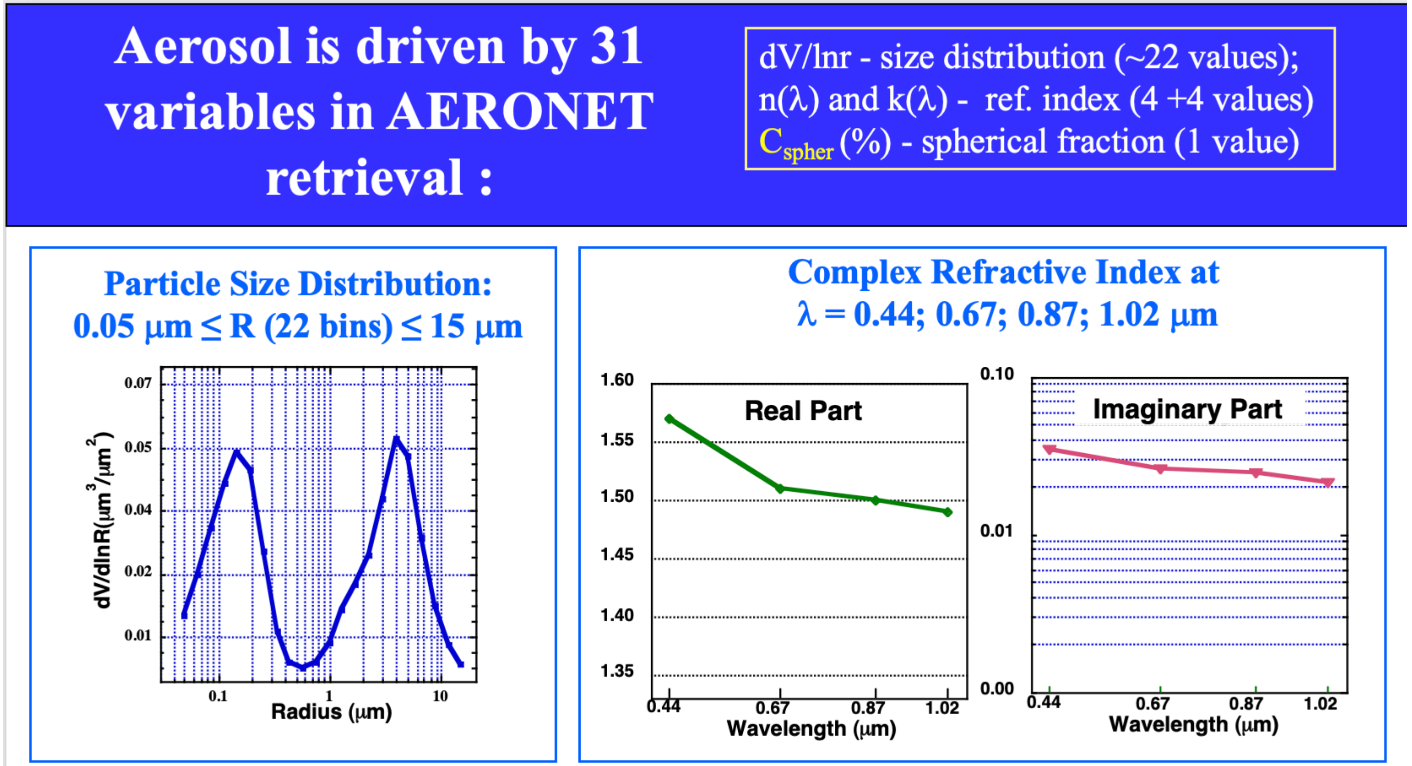
Aerosol components and bins (tracers)

	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 Hydro philic**	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 representation in *Remote Sensing*







GRASP: **G**eneralized **R**etrieval of **A**tmosphere and **S**urface **P**roperties

GRASP is advanced algorithm for retrieval of aerosol, gas and surface properties from diverse remote sensing observations and any combination of them based on:

Forward Model for rigorous simulation of atm. radiation.

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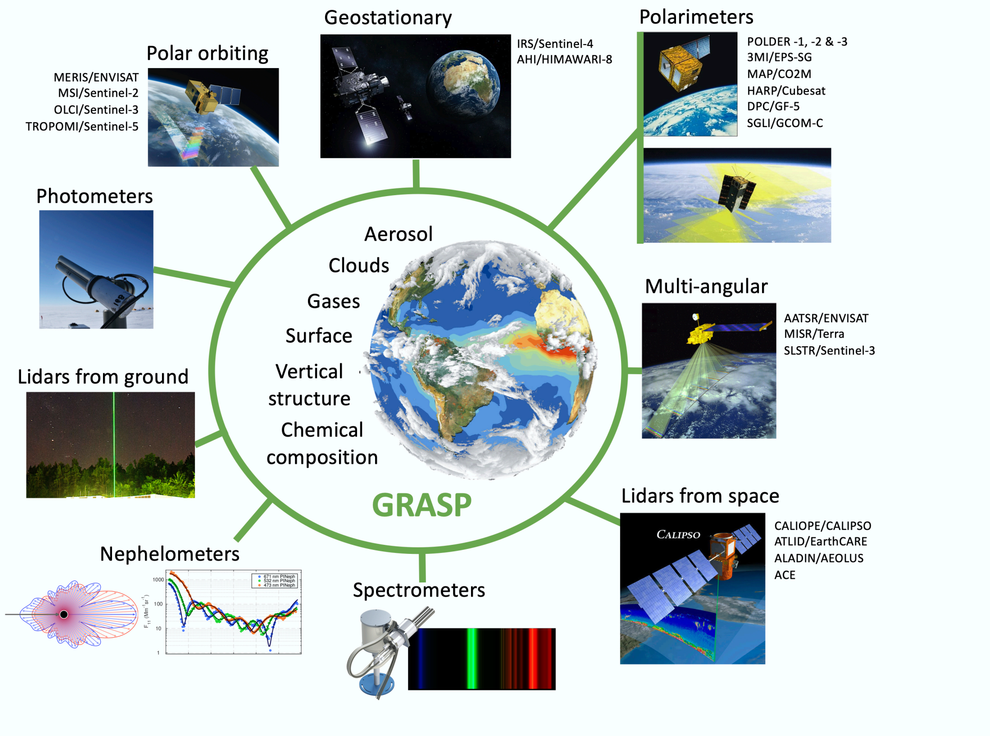
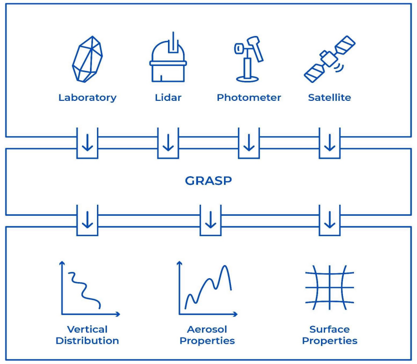
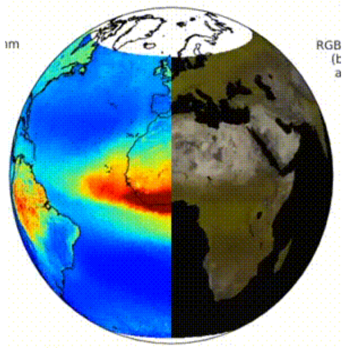
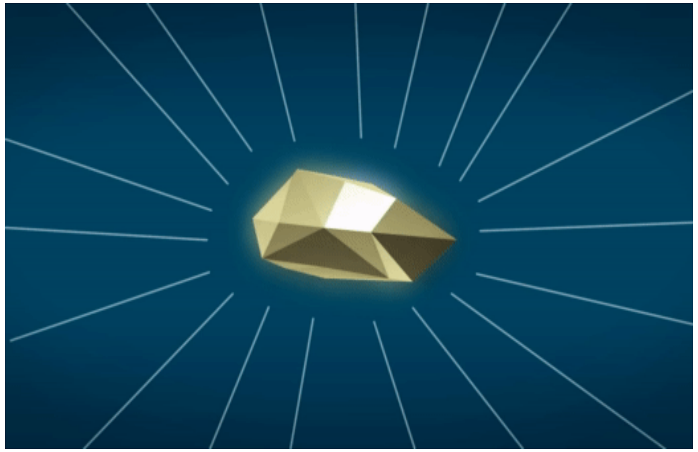
Inversion with *ap*plying multiple *a priori* constraints



Dubovik et al. "A Comprehensive Description of Multi-Term LSM for Applying Multiple a Priori Constraints in Problems of Atmospheric Remote Sensing: GRASP Algorithm, Concept, and Applications", Front. Remote Sens., 2021







GRASP: Generalized Retrieval of Atmosphere and Surface Properties

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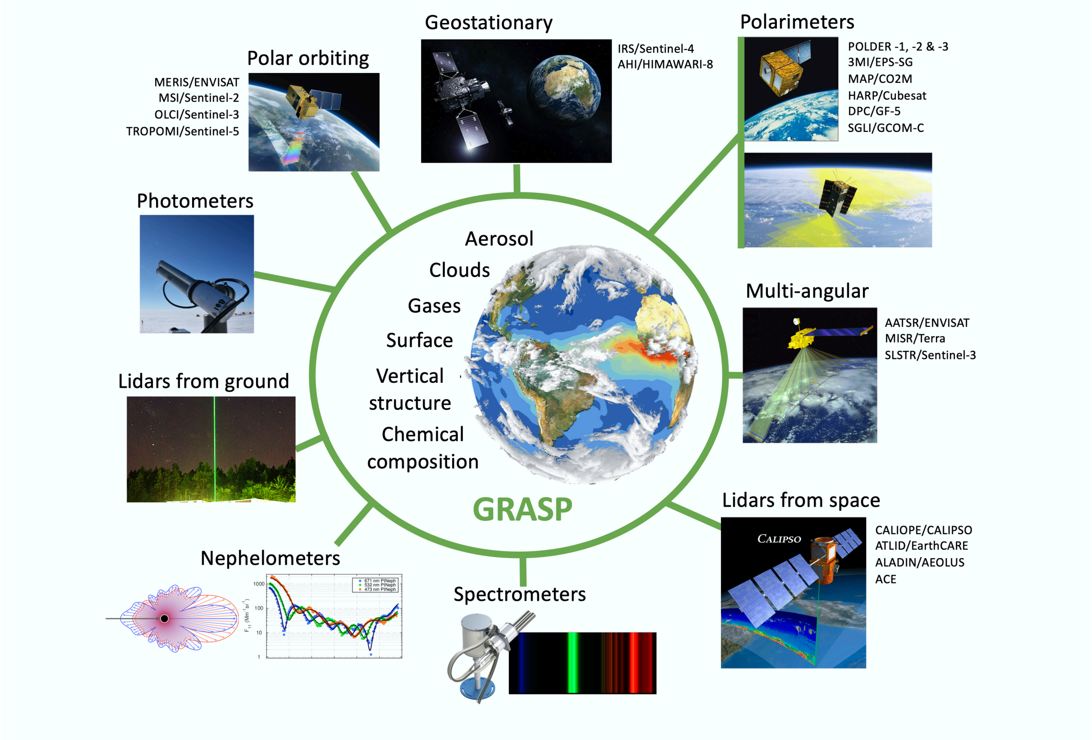

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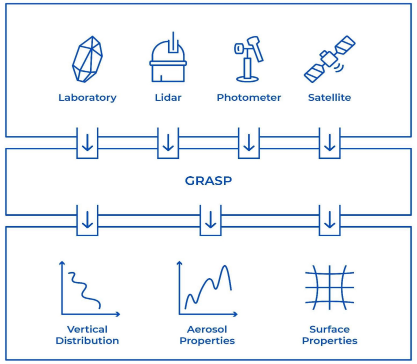
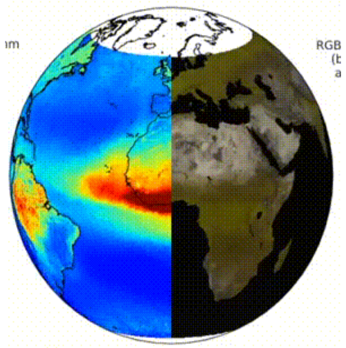
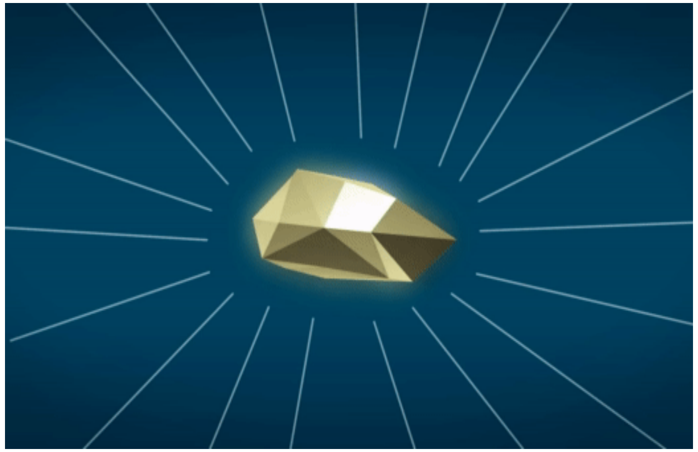
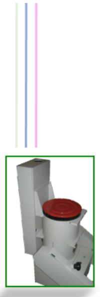
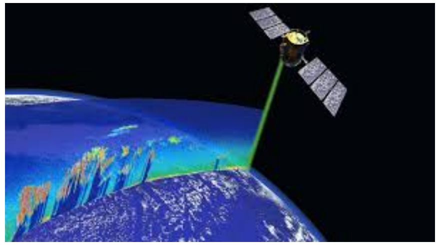






Angular aspects of different observations

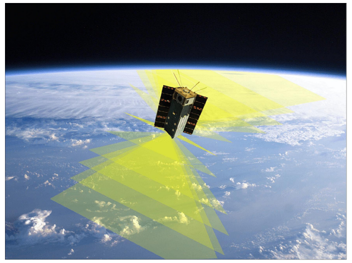




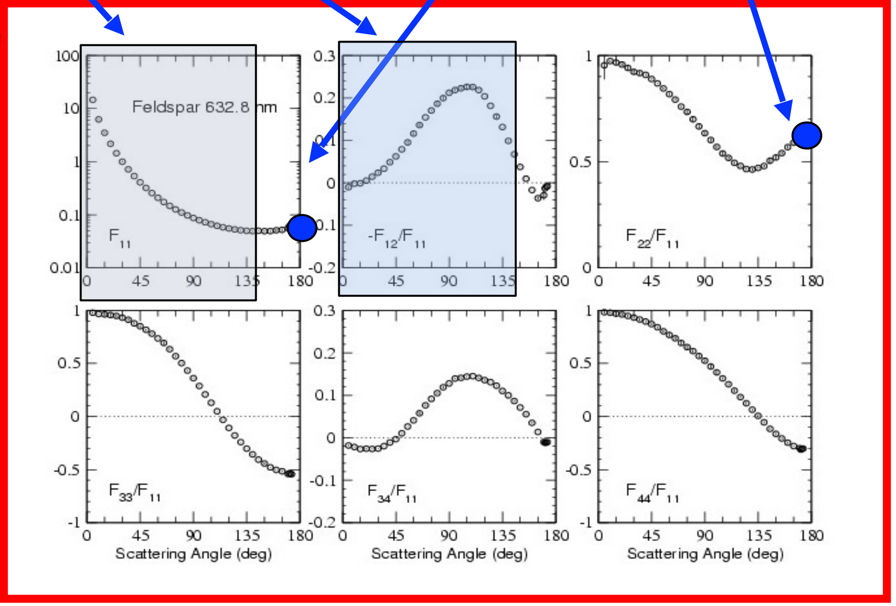
Lidar - active

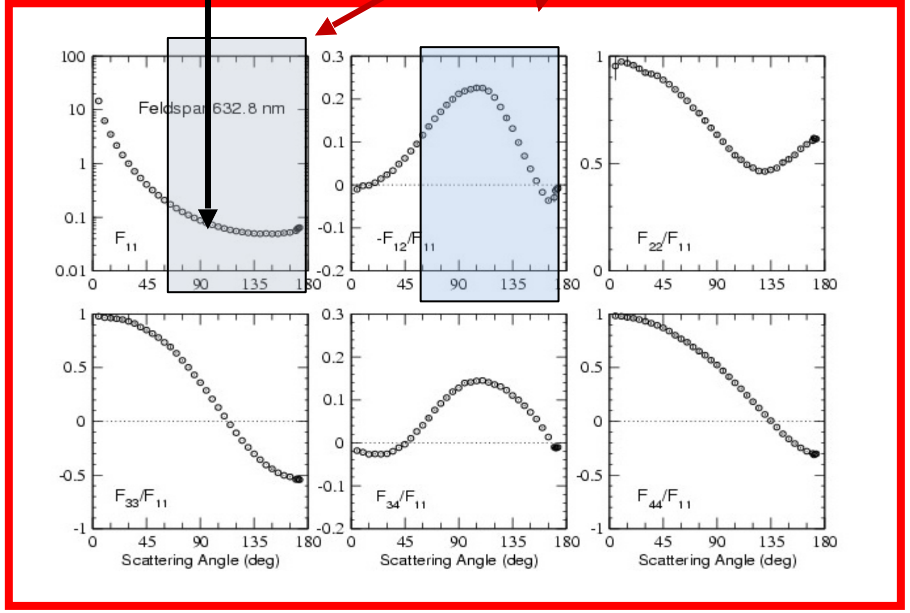
Single-view radiometer

Multi-angular polarimeter



Neph-elometer





P_{11} – intensity, P_{12} , P_{22} - state of **polarization**

size, shape, absorption, refraction

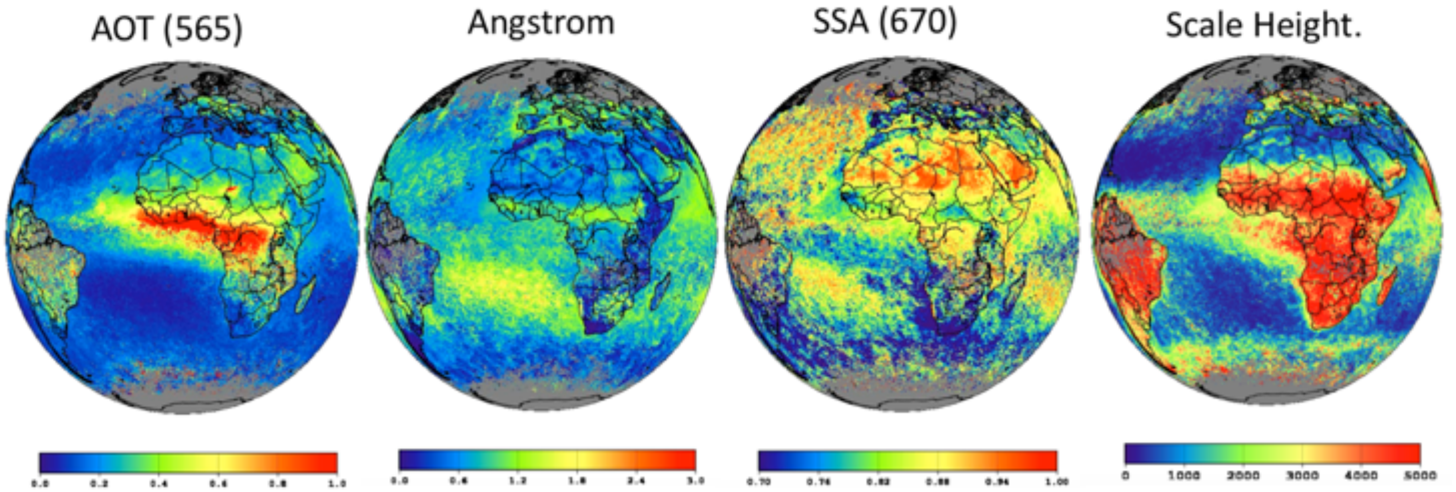
$$\begin{pmatrix} I_s \\ Q_s \\ U_s \\ V_s \end{pmatrix} \propto \begin{pmatrix} P_{11}(\Theta) & P_{12}(\Theta) & 0 & 0 \\ P_{12}(\Theta) & P_{22}(\Theta) & 0 & 0 \\ 0 & 0 & P_{33}(\Theta) & P_{34}(\Theta) \\ 0 & 0 & -P_{34}(\Theta) & P_{44}(\Theta) \end{pmatrix} \begin{pmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{pmatrix}$$

POLDER/PARASOL 2004-2013 4 products

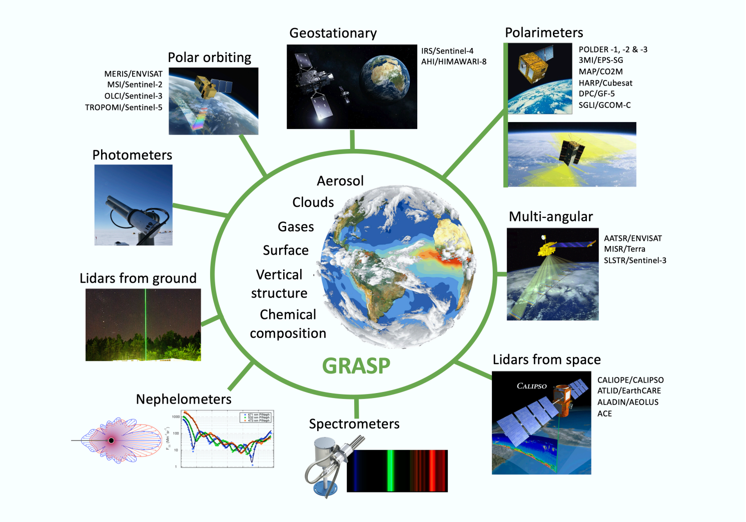
Chen et al., 2020
Li et al., 2019
Zhang et al. 2021
Dubovik et al. 2021

Winter 2009

Ocean and land



GRASP



<u>AEROSOL</u>: AOD spectral, AOD fine/coarse, Angstrom, SSA, AAOD, aerosol height, spectral complex index of refraction, sphericity fraction.

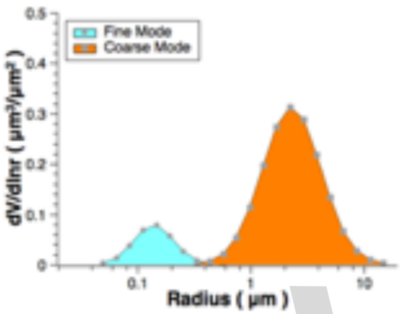
<u>SURFACE</u>: land BRDF spectral, BPDF spectral;
ocean wind speed and water leaving radiances, etc.

Important features of GRASP retrieval:

- Globally the same initial guess for aerosol;
- Globally the same set of a priori constraints;
- No location specific assumptions;
- Retrieval on 6 km resolution, no averaging;
- Surface retrieved simultaneously

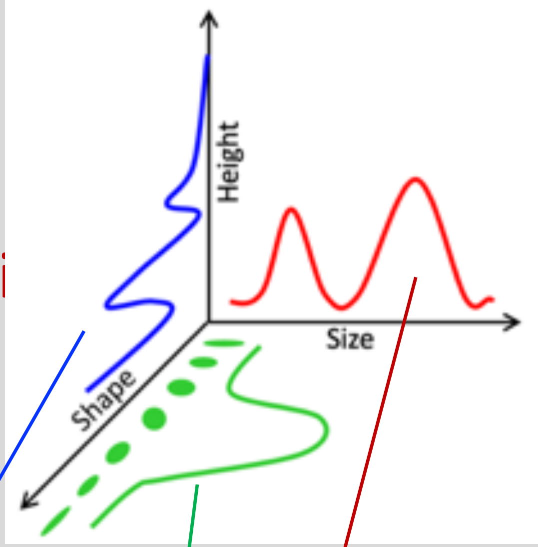
Aerosol model in GRASP:

Multi-component mixture of spheres and randomly oriented spheroids



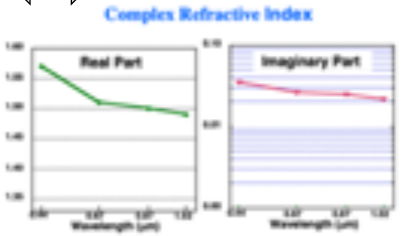
TO be RETRIEVED
OR
ASSUMED

Size.
Shape
Vertical
distributi



Chemical composition
Internal mixture

n(λ), k(λ)



Measurements:

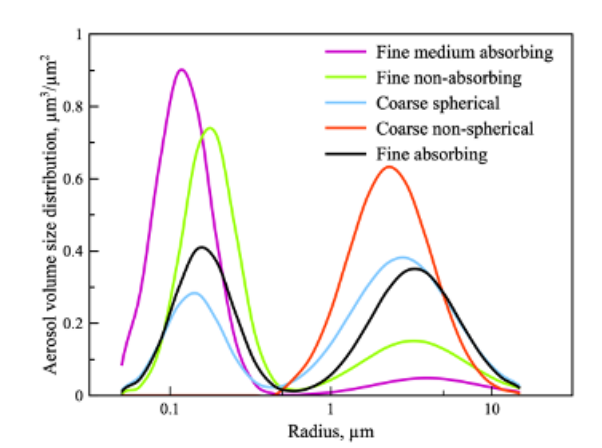
$$\tau_{scat/ext} = \sum_{k=1}^{K}\left(\int_{h_{min}}^{h_{max}} \int_{\varepsilon_{min}}^{\varepsilon_{max}} \int_{r_{min}}^{r_{max}} \frac{C_{scat/ext}\left(n_k(\lambda);k_k(\lambda);h;\varepsilon;r\right)}{V(r)} \frac{dV_k(h)}{dh}\frac{dn_k(r)}{d\varepsilon}\frac{dV_k(r)}{dr}\, dh\, d\varepsilon\,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

Aerosol modeling approaches in GRASP

1. GRASP 5 aerosol models approach

(Lopatin et al., AMT, 2021)

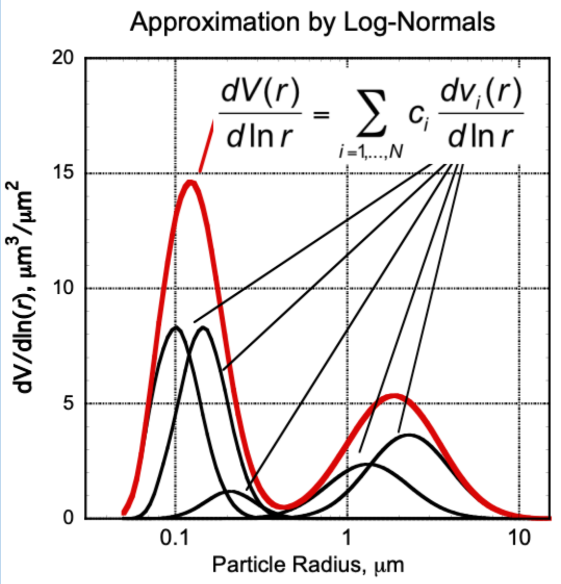
1. Aerosol component concentration
2. Total concentration



2. GRASP 5LN bins: Full Microphysics approach

(Dubovik et al.,2011, 2021)

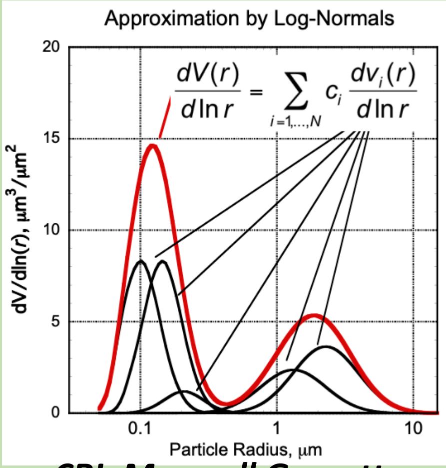
1. 5 LNBins
2. Total Concentration
3. Non-spherisity
4. Spectral CRI



3. GRASP 5LN Chemical Component approach

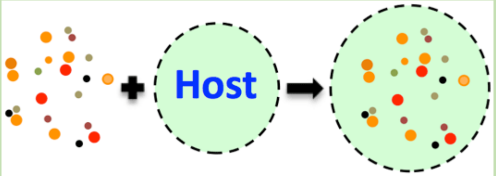
(L. Li et al., ACP, 2019)

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



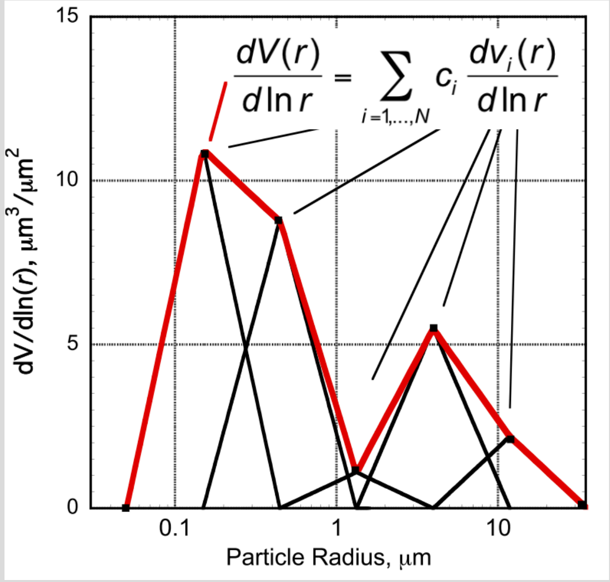
CRI: Maxwell Garnett

effective medium approximation



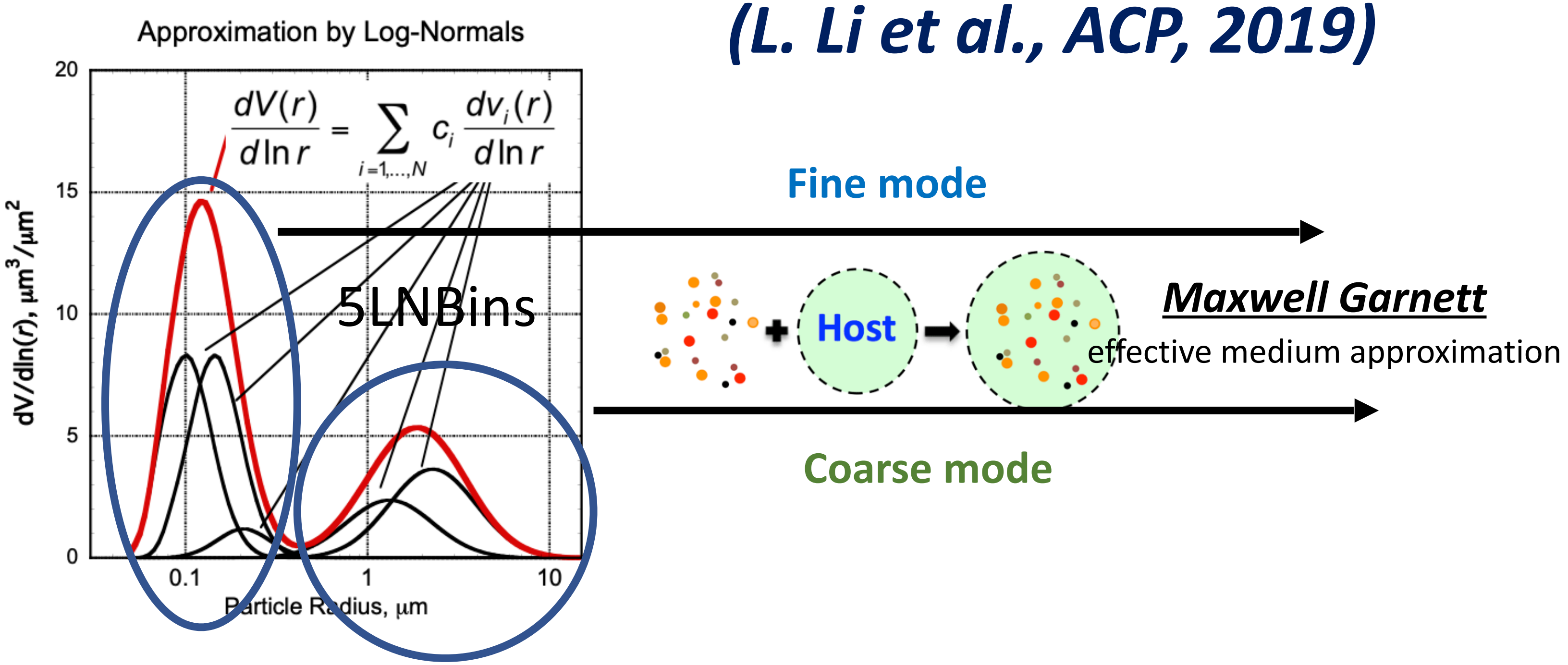
4. GRASP 22 bins: Full Microphysics approach

1. 22 Triangular bins
2. Total Concentration
3. Non-spherisity
4. Spectral CRI



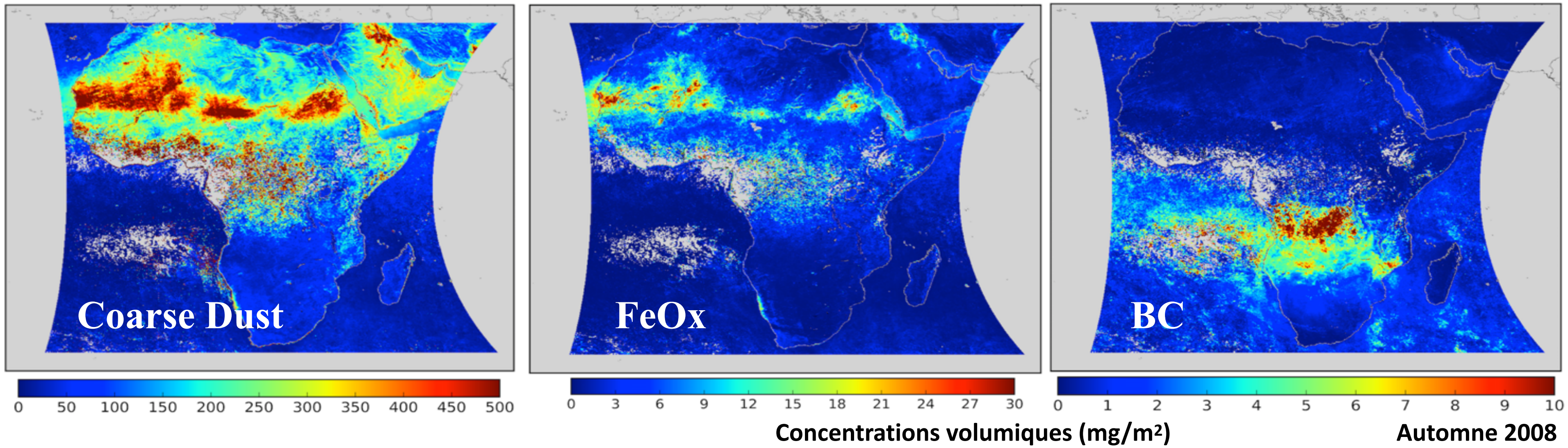
Evolution: GRASP Component approach

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



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

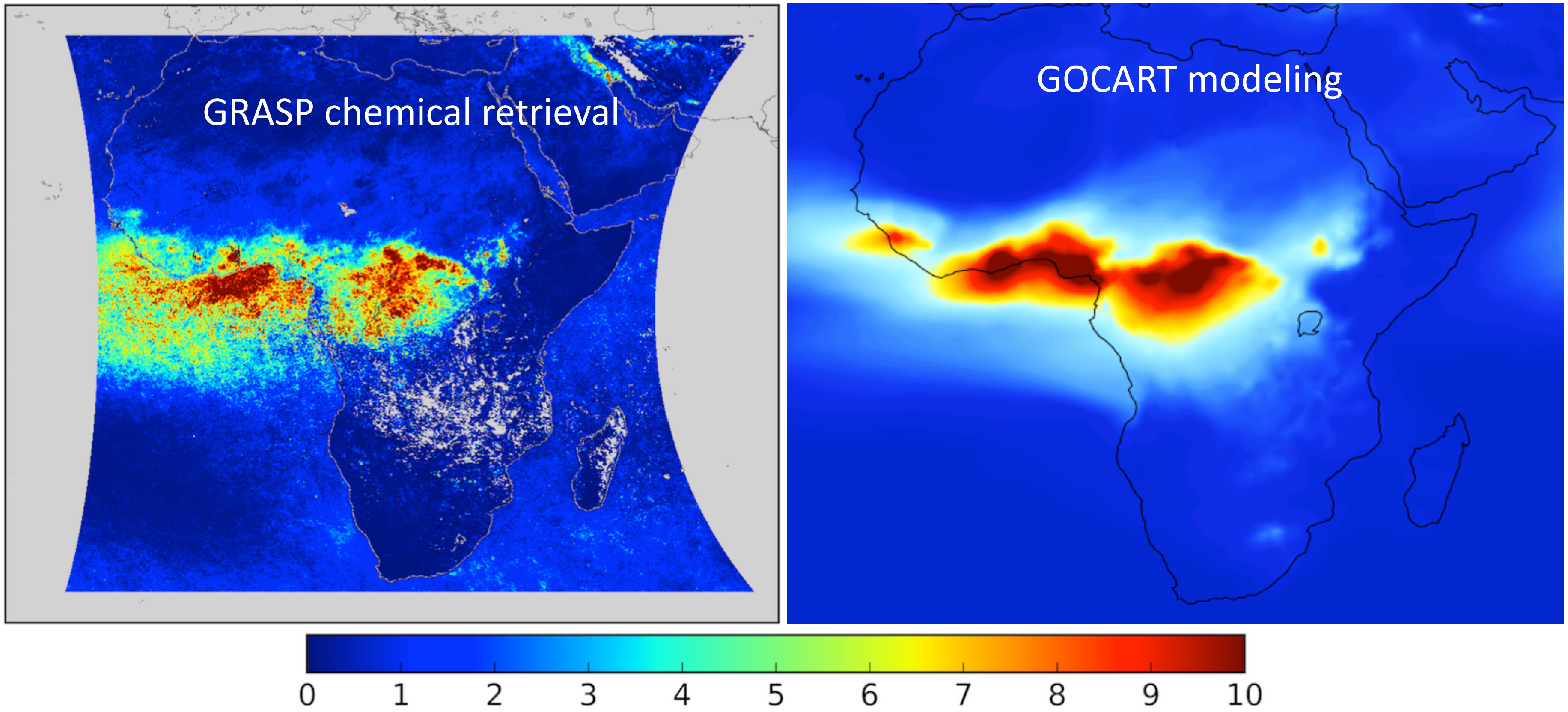
Absorbing insoluble (FeOx)
Non-absorbing insoluble (Dust)
Non-absorbing soluble (SS, etc.)
Water
Coarse mode



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

Example of chemical composition retrieval

Black Carbon mass concentration(mg/m²) in January 2008



Details are in Li et al. (2019)

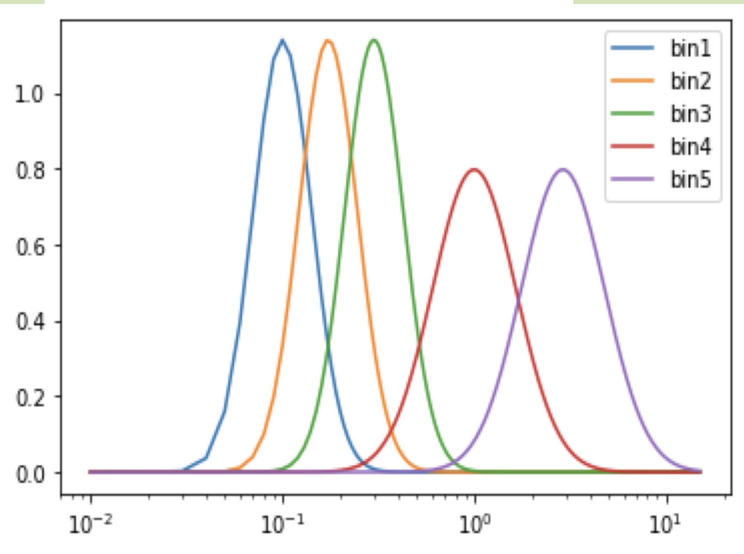
Emission retrievals –Chen et al. (2018, 2019, 2022)

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

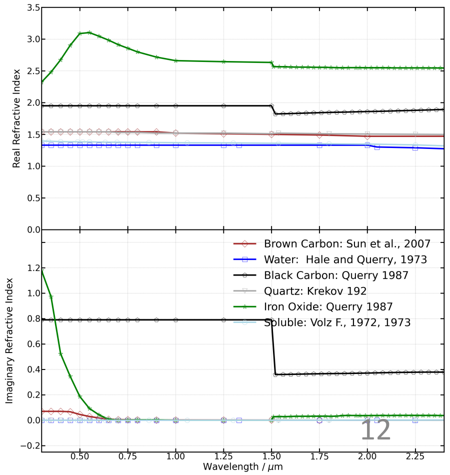
Effective refractive index:

Fine mode :

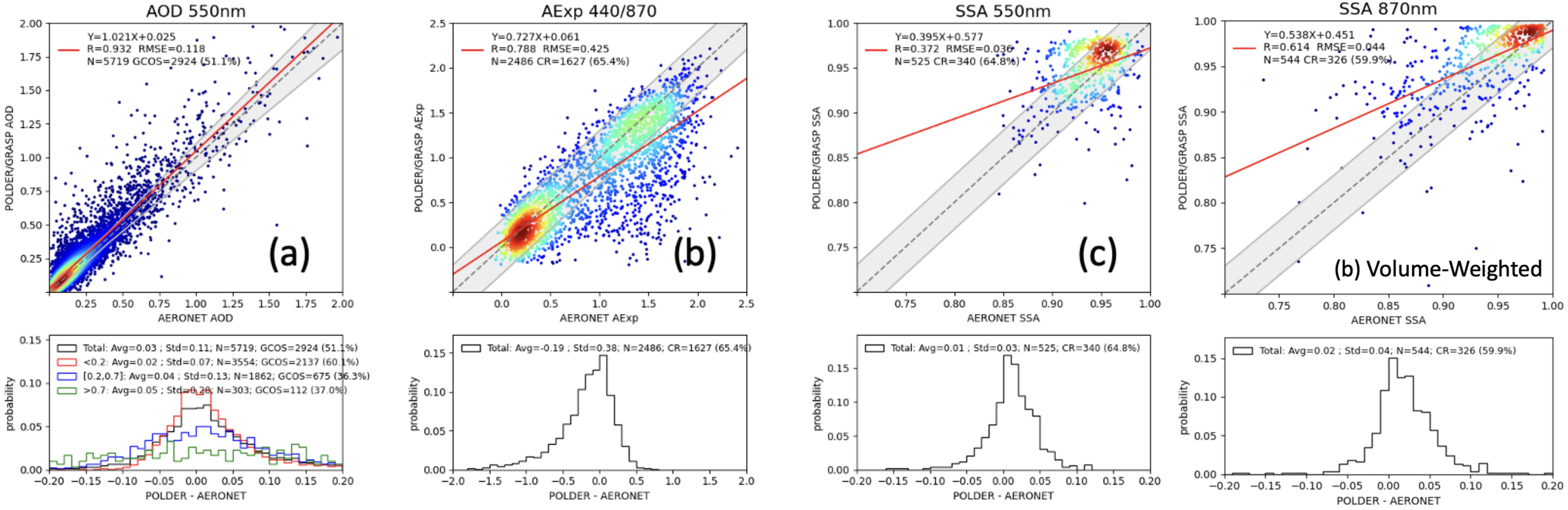
$$\hat{m}_{eff}^{fine} = \hat{m}_{BC}\delta_{BC}^{phil} + \hat{m}_{BrC}\delta_{BrC} + \hat{m}_{SU}\delta_{SU} + \hat{m}_{Water}\delta_{Water}^{fine}$$

Coarse mode:

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

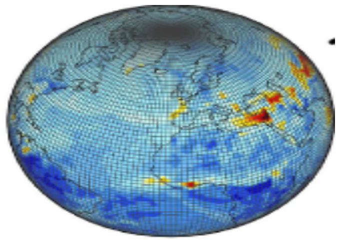


Baseline GRASP **2 modes** approach PARASOL retrievals



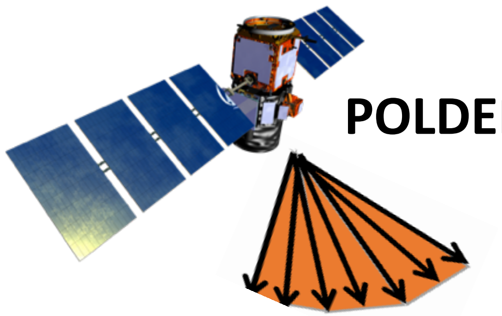
TEST-1: Nature run retrieval

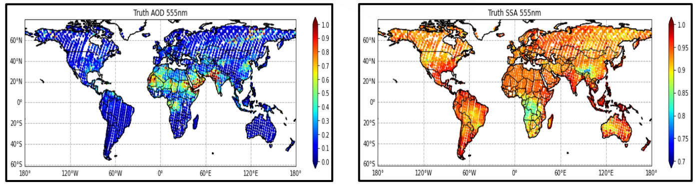
MERA-2/CAMS Global chemistry-transport model



NATURE RUN
or *pseudo-reality*
BC, OM, SU, Sea Salt, Dust

4-D - mixing ratios;
Variability: time, vertical, horizontal

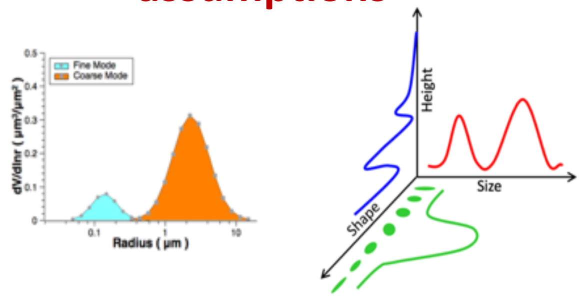




Simulation of POLDER-3 measurements

GRASP

Different aerosol assumptions



Retrieval aerosol properties

« reality »

Aerosol columnar properties
AOD, AE, SSA, etc.

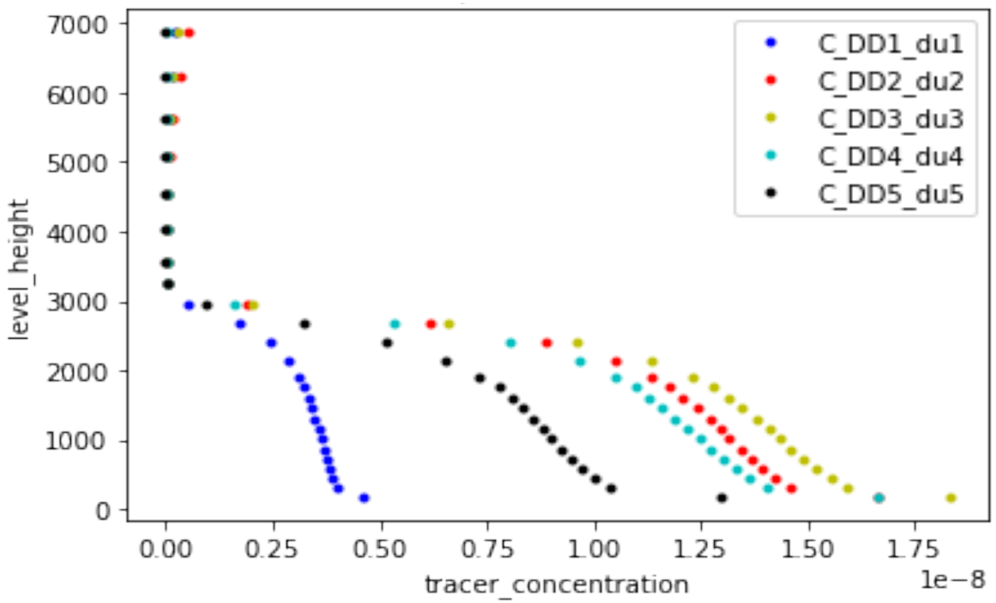
retrieval

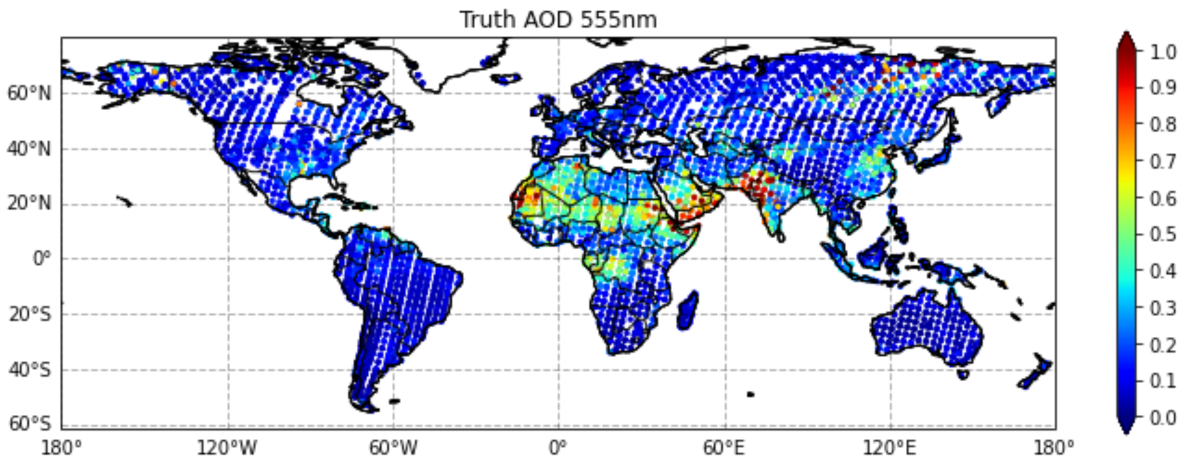
Comparison
« reality » vs retrieval

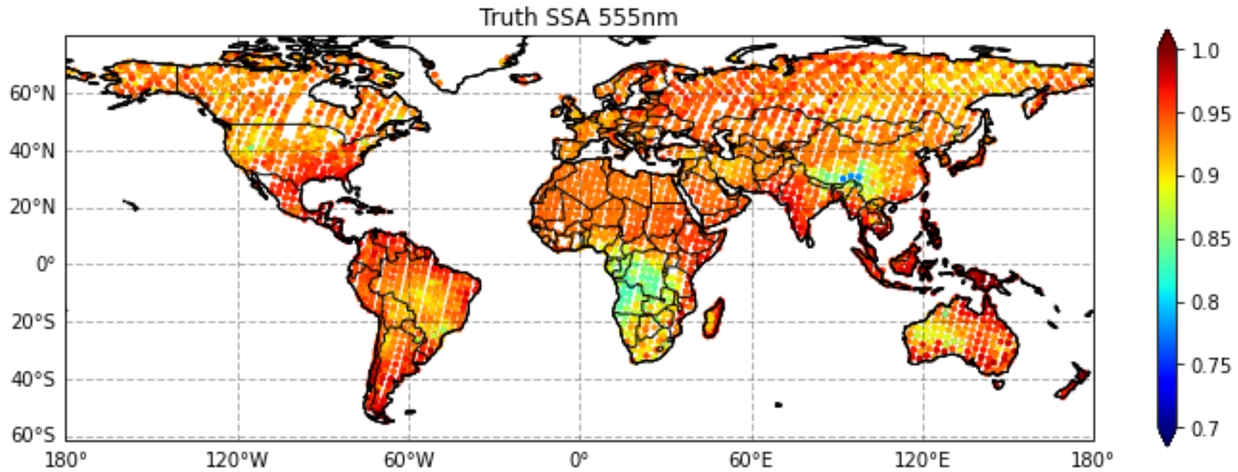
Synthetic dataset

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)





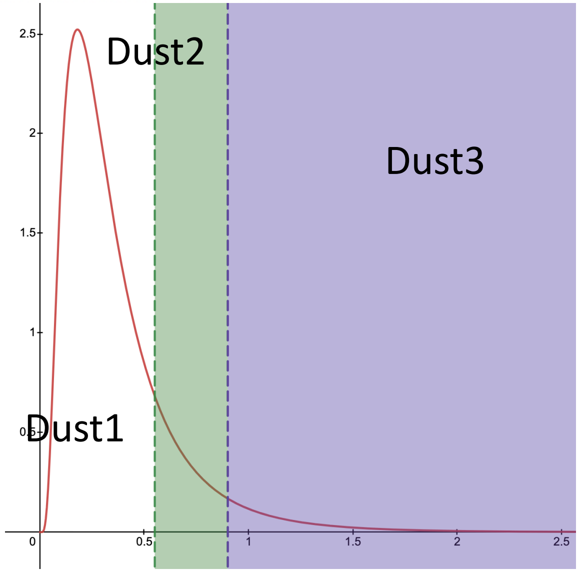


Size distribution in CAMS/MERRA-2 versus GRASP

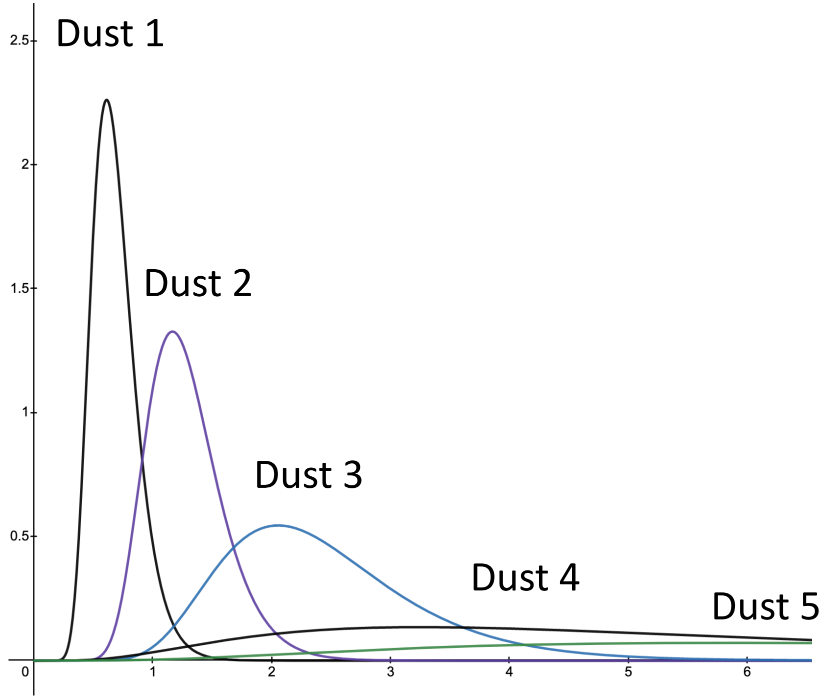
- The way of cutting the size distribution to define bins for Dust and Sea Salt in CAMS looks very different from the one used in remote sensing retrieval where SD is smooth function.

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

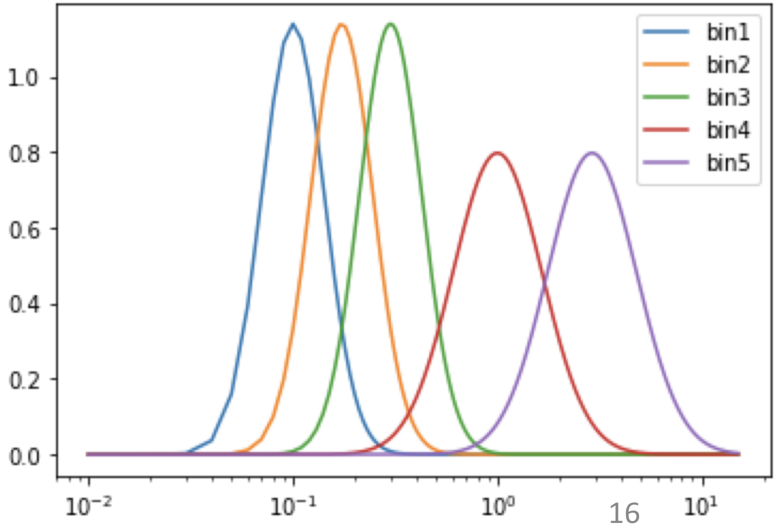
$$n(r) = \frac{\mathrm{d}N(r)}{\mathrm{d}r} = \frac{N}{\sqrt{2\pi}\, r\ln(\sigma)} \exp\left(-\frac{\ln^2(r/r_{\mathrm{mod}})}{2\ln^2(\sigma)}\right)$$



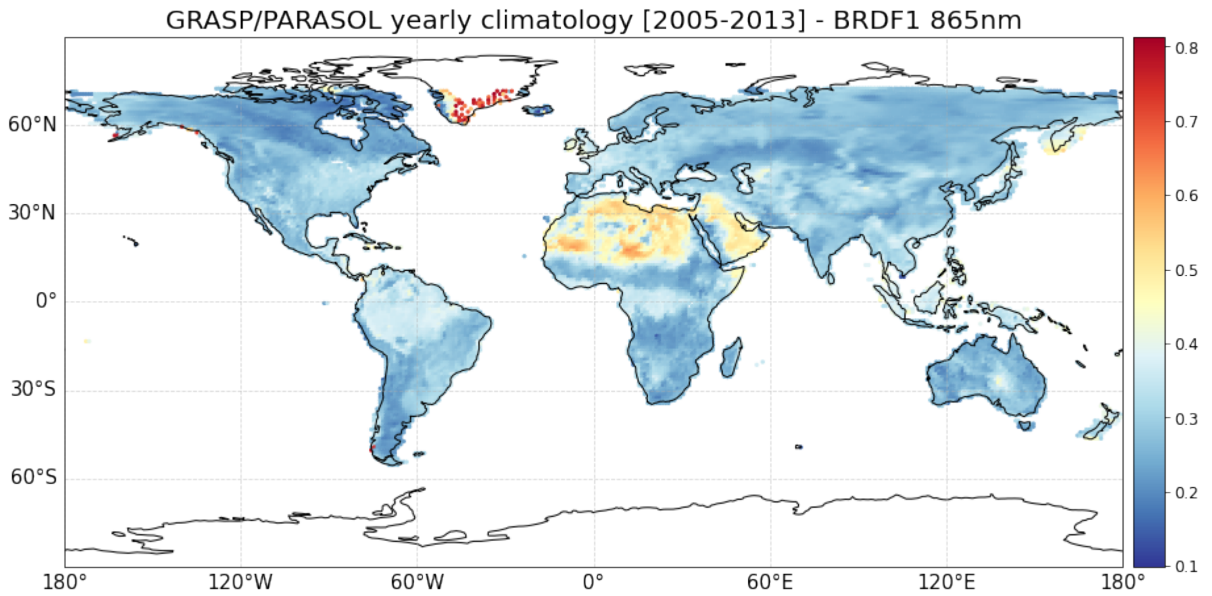
Dust tracer from MERRA2 Natural Run

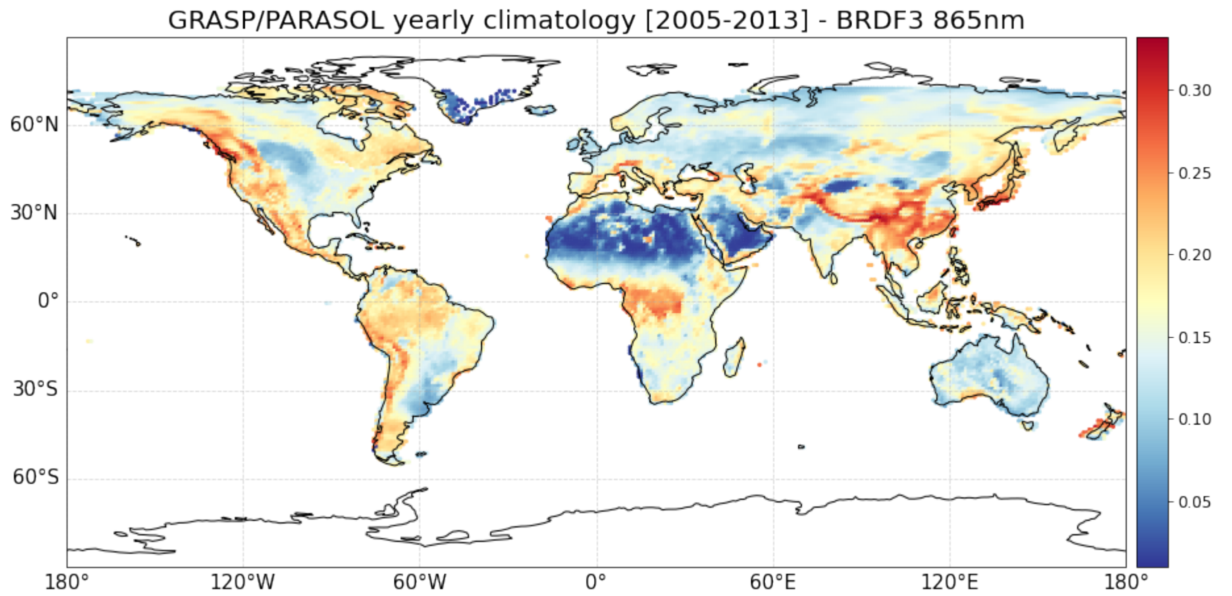


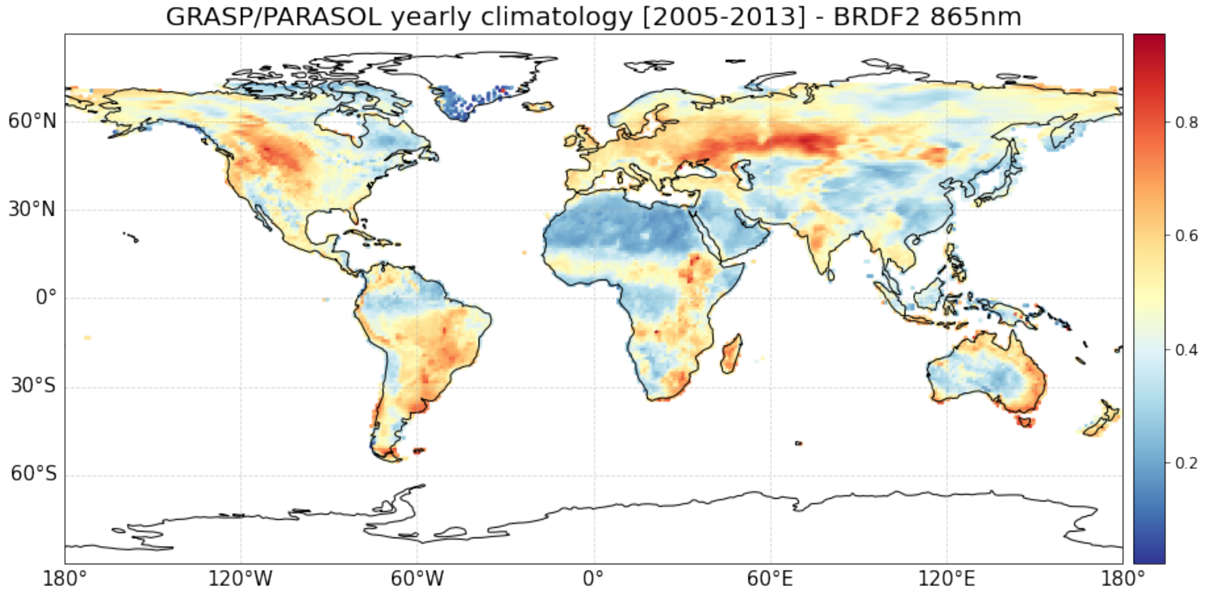
GRASP 5 LN SD bins

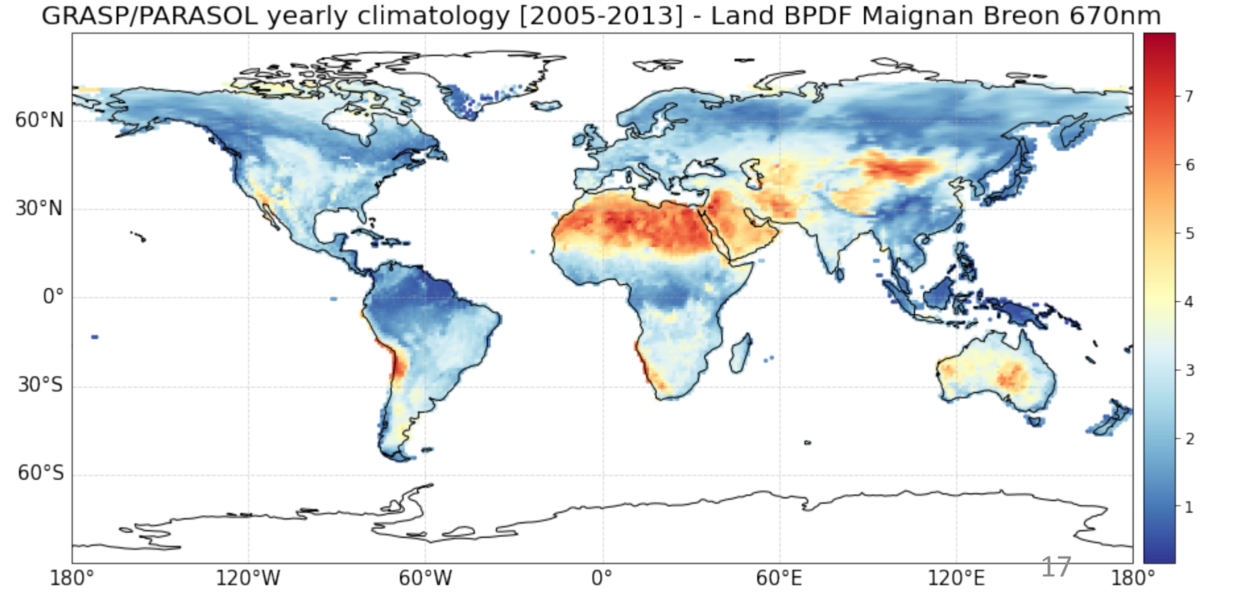


Surface simulation: BRDF and BPDF



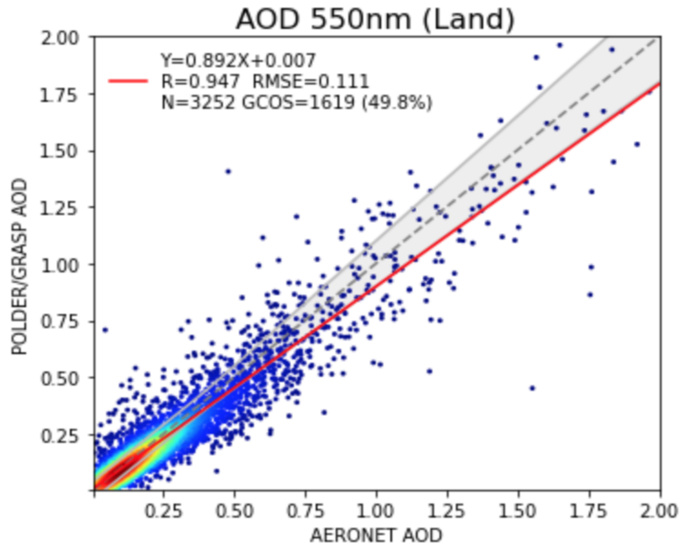


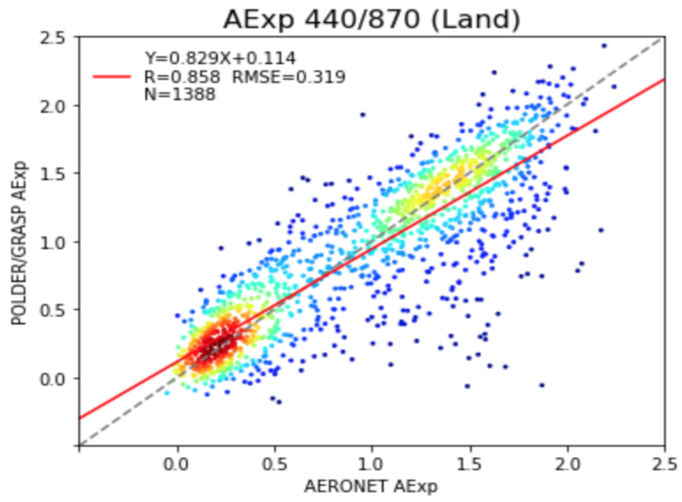




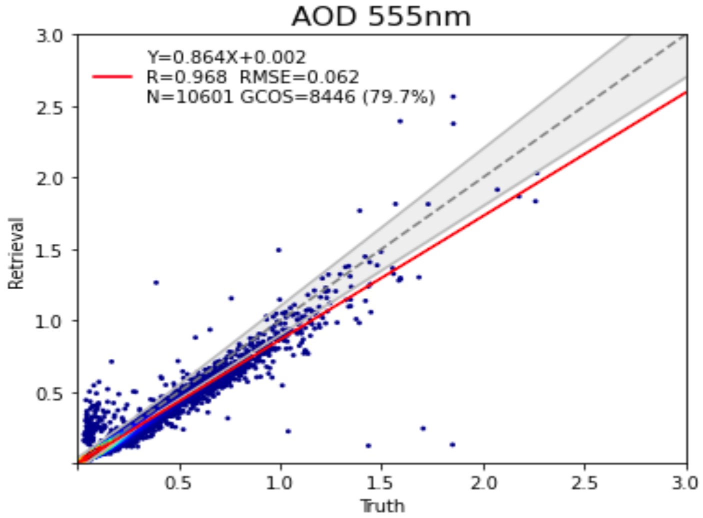
GRASP/Component approach performance

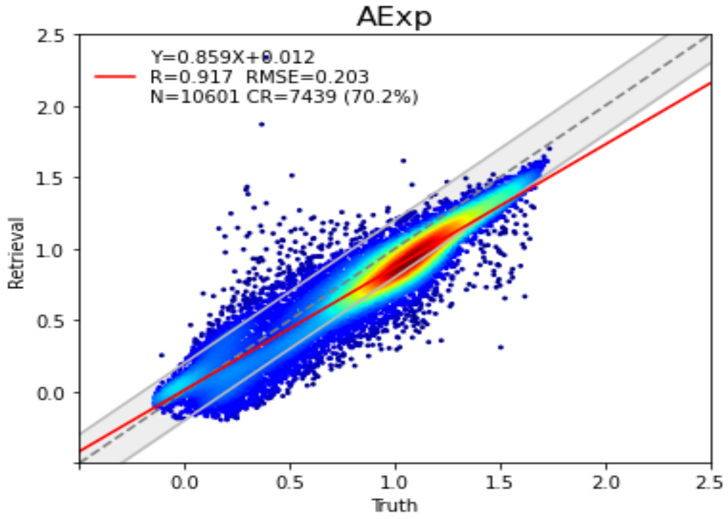
POLDER-3 retrieval (2008)





MERA-2 synthetic data inversion

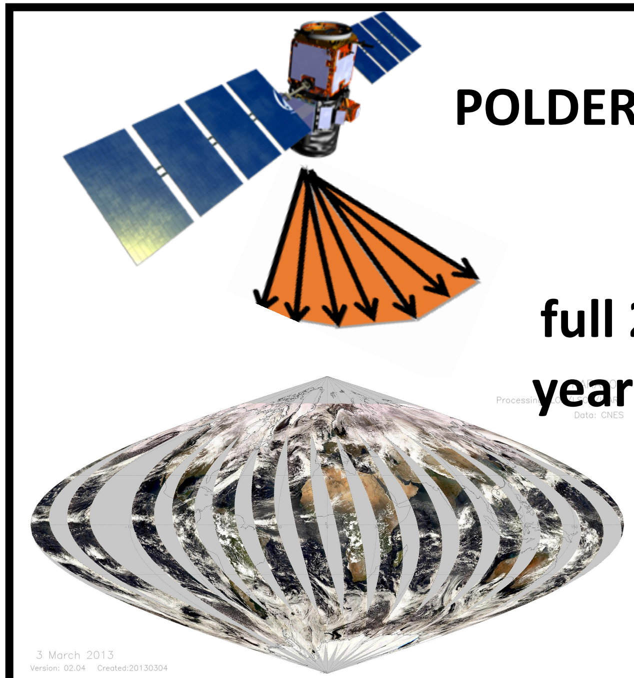




Over land

Remote sensing approach works well on synthetic data

TEST-2: reality test

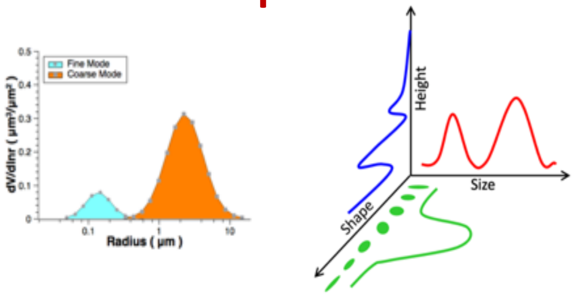


POLDER-3

full 2008 year data

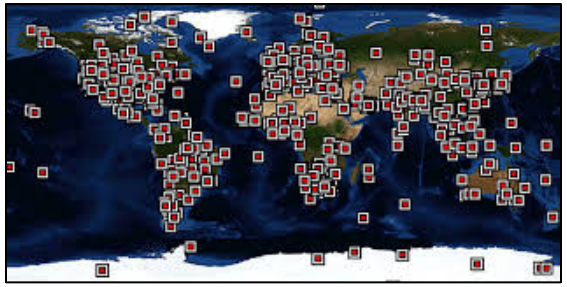
GRASP

Different aerosol assumptions



Retrieval aerosol properties

retrieval from REAL data



AERONET

« reality »

Aerosol columnar properties

AOD, AE, SSA, etc.

retrieval

Comparison

« reality » vs retrieval

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

GRASP / remote sensing

2 modes

Optical properties

(*refractive indices*)

≠

CAMS / MERRA-2

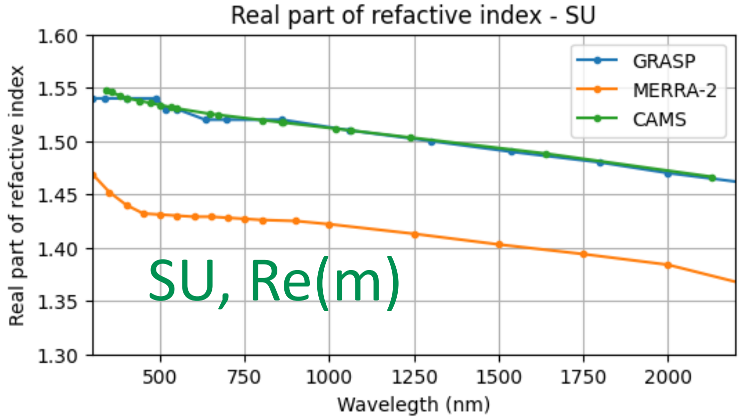
5 modes

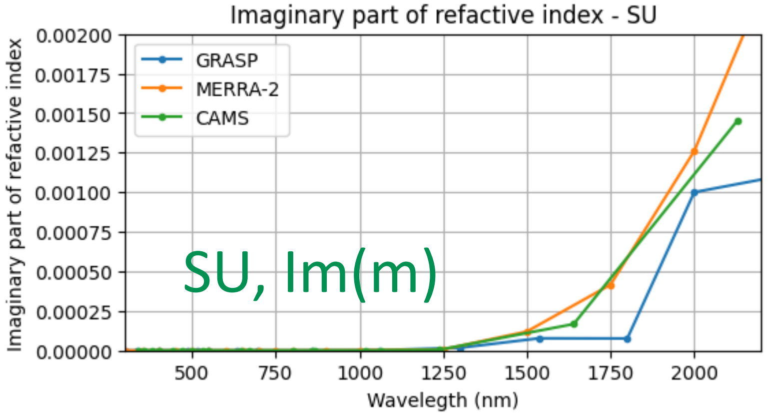
Optical properties

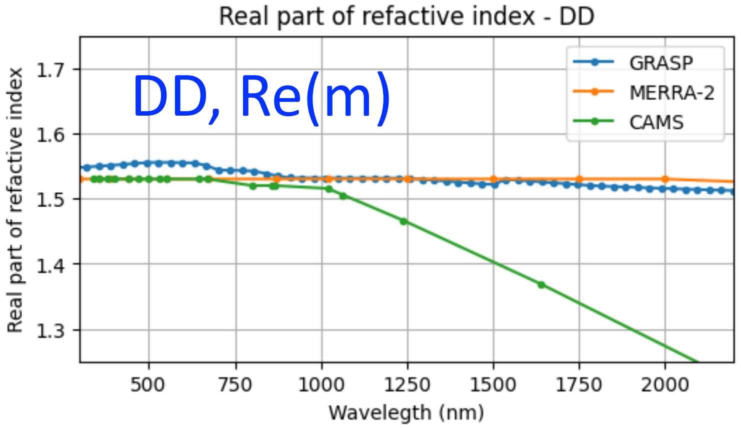
(*refractive indices*)

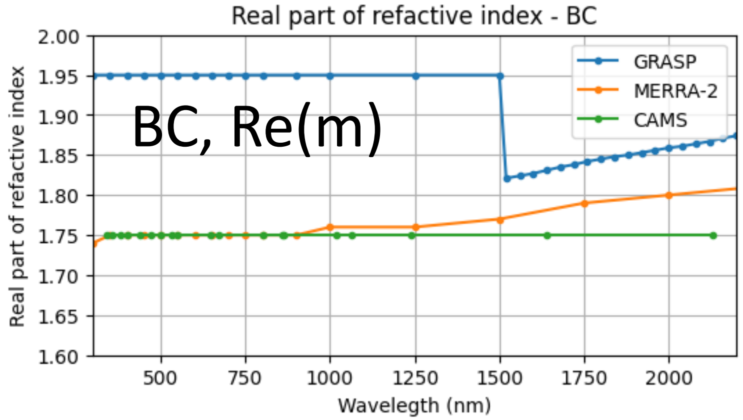
Feasibility tests	Harmonization toward CAMS aerosol representation
1	- Adjustment of the **complex refractive** index of GRASP aerosol chemical components;
2	- **Sea Salt** and **Dust** in separates modes
3	- **Sea Salt** and **Dust** in separates modes

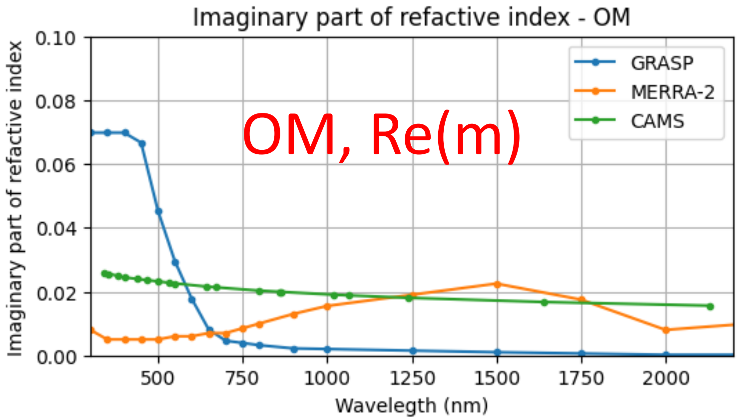
Refractive indices for aerosol species

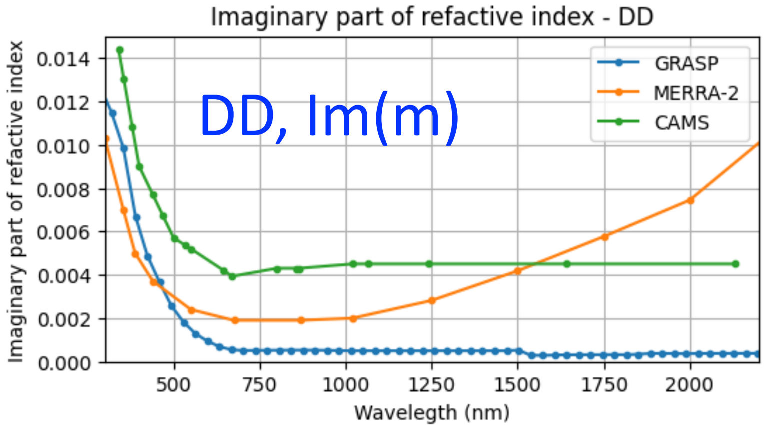


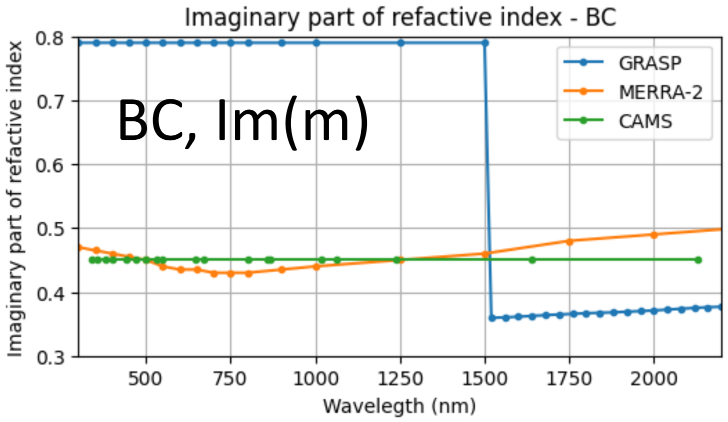


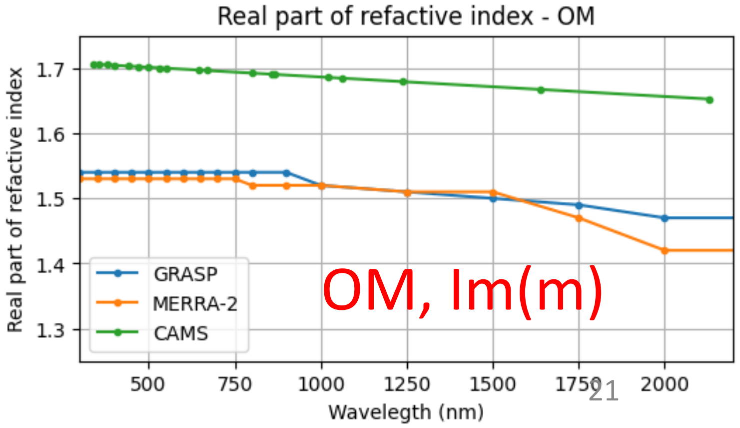






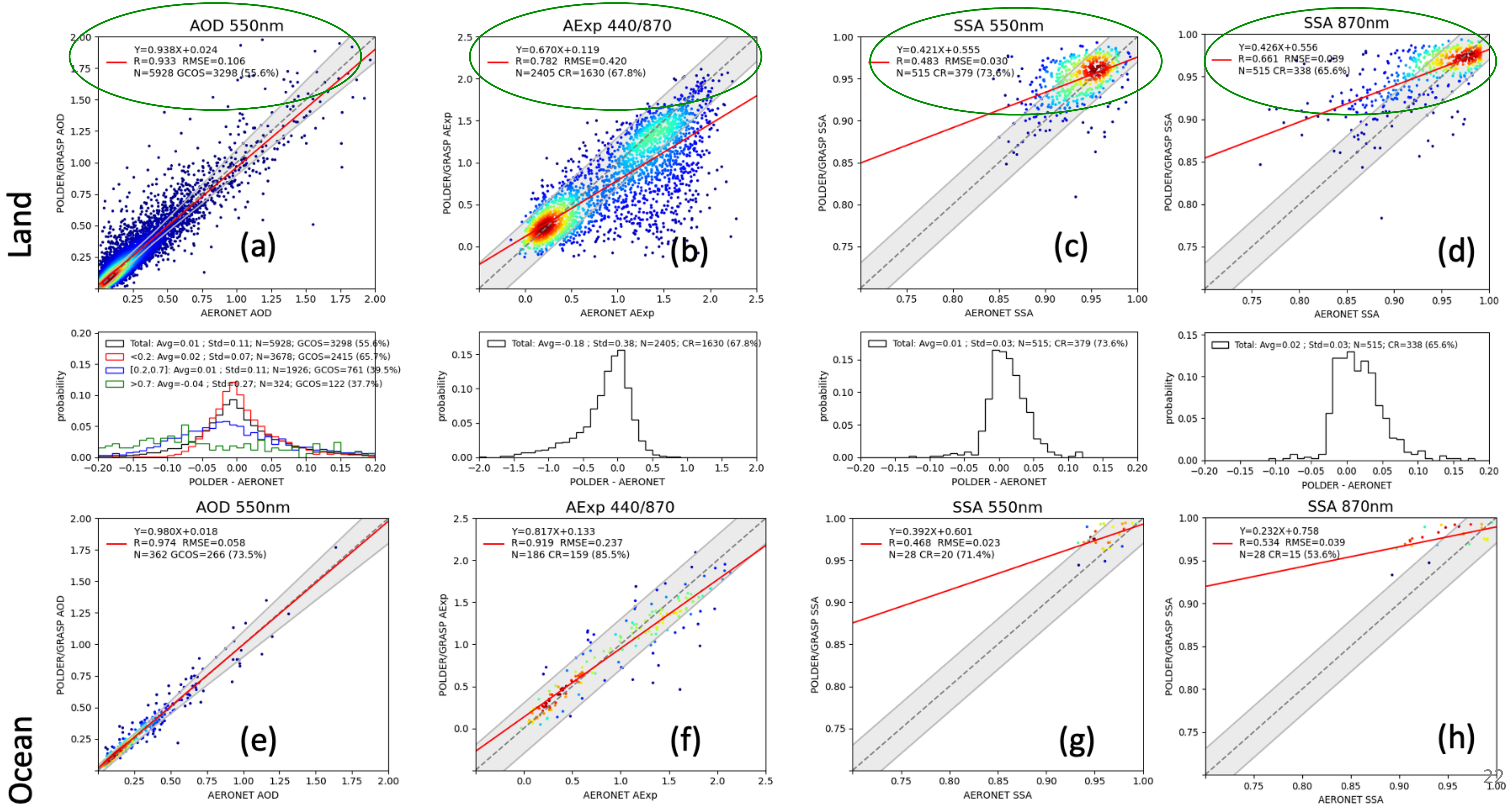




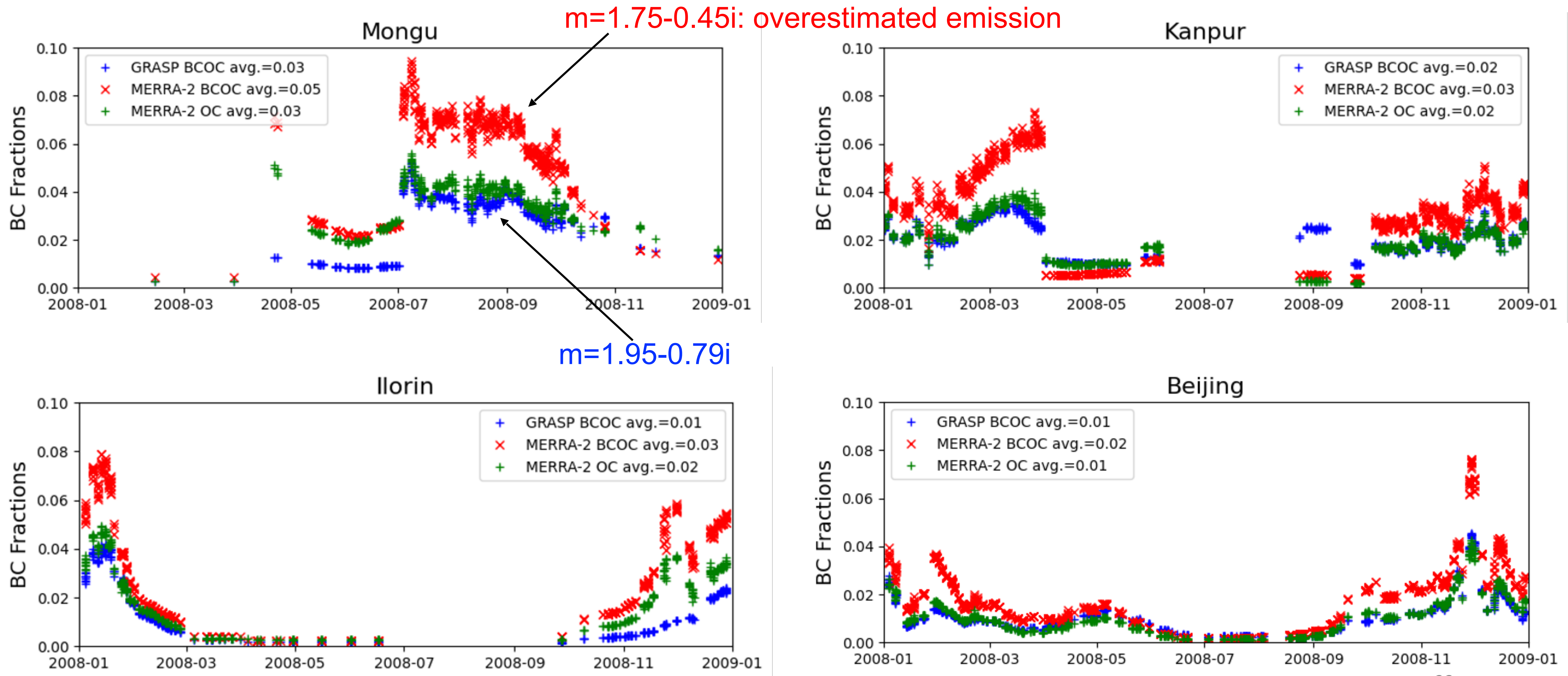


Refractive index harmonization: MERRA-2/CAMS BC and OM refractive index in GRASP

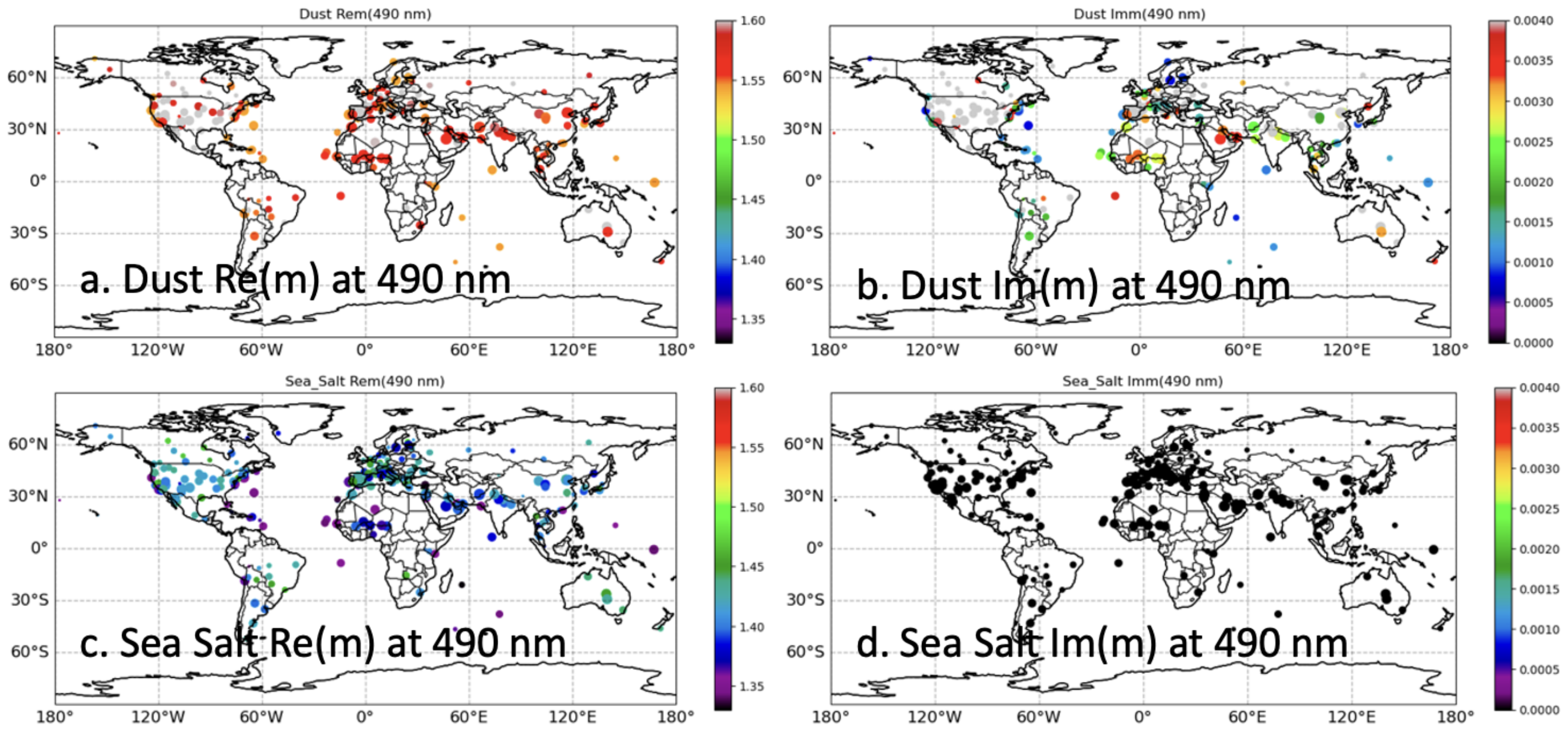
Similar performance

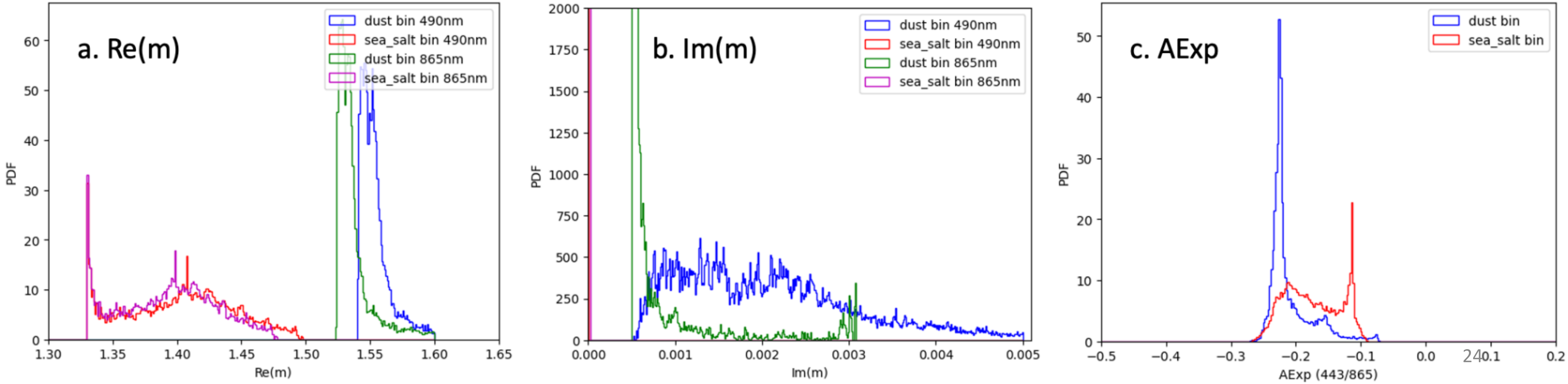


Refractive index harmonization: MERRA-2/CAMS BC and OM refractive index in GRASP



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:

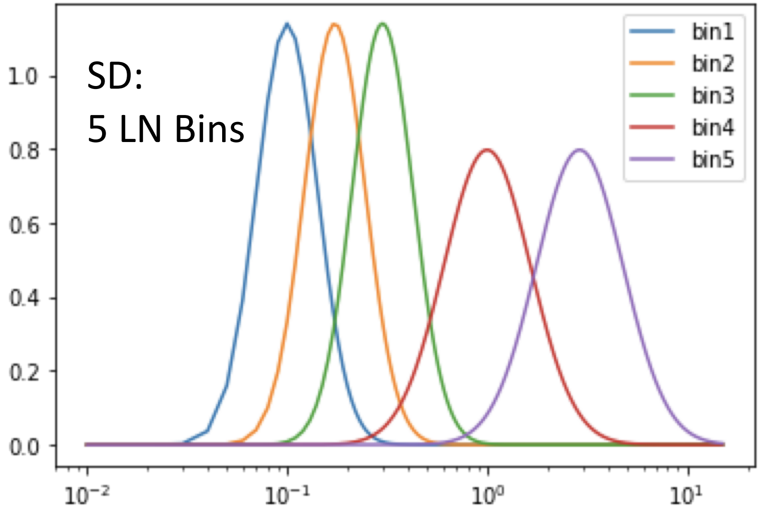
Feasibility tests with real PARASOL data	Performance in **AOD**	Performance in **AE**	Performance in **SSA**
Adjustment of the complex refractive index of aerosol	Same quality	Same quality	Same quality

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;

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:

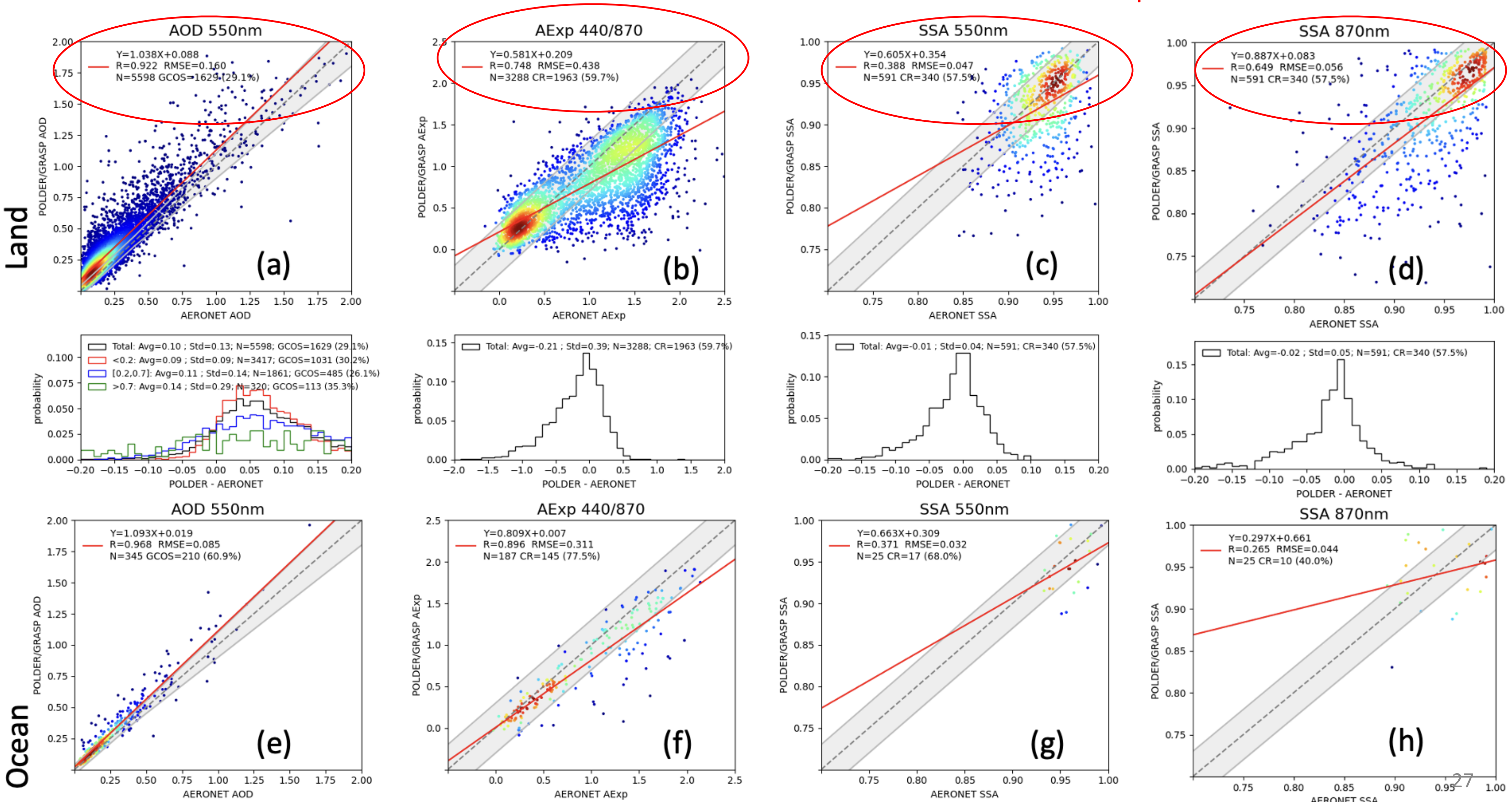
$$\hat{m}_{eff}^{fine_2} = \hat{m}_{BC}\delta_{BC}^{phil} + \hat{m}_{BrC}\delta_{BrC}^{phil} + \hat{m}_{SU}\delta_{SU} + \hat{m}_{Water}\delta_{Water}^{fine}$$

Coarse mode:

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

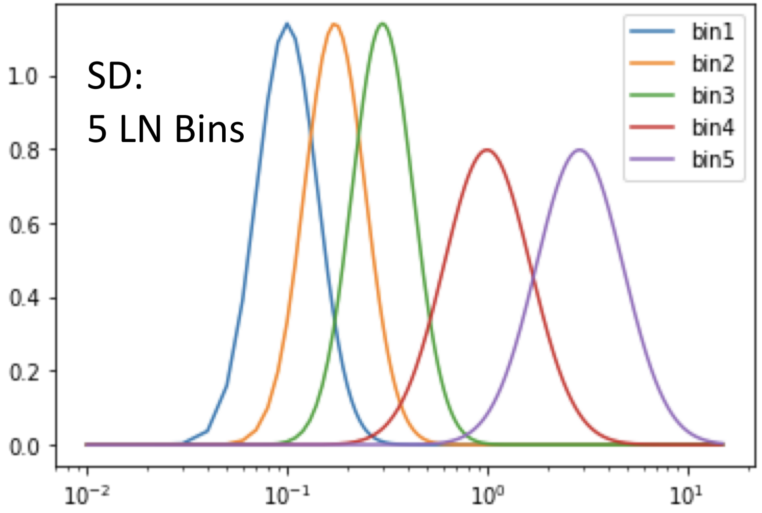
3 aerosol modes: Hydrophilic and Hydrophobic **BC and BrC** in separate modes (performance on PARASOL measurements)

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	Dust: Quarts
Fine mode	Bin1, Bin2	✔	✔	✔	✔	✔			
Coarse mode 1	Bin2, Bin3	✔				✔	✔		
Coarse mode 2	Bin4, Bin5	✔						✔	✔



Effective refractive index:

Fine mode:

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

Fine mode 2:

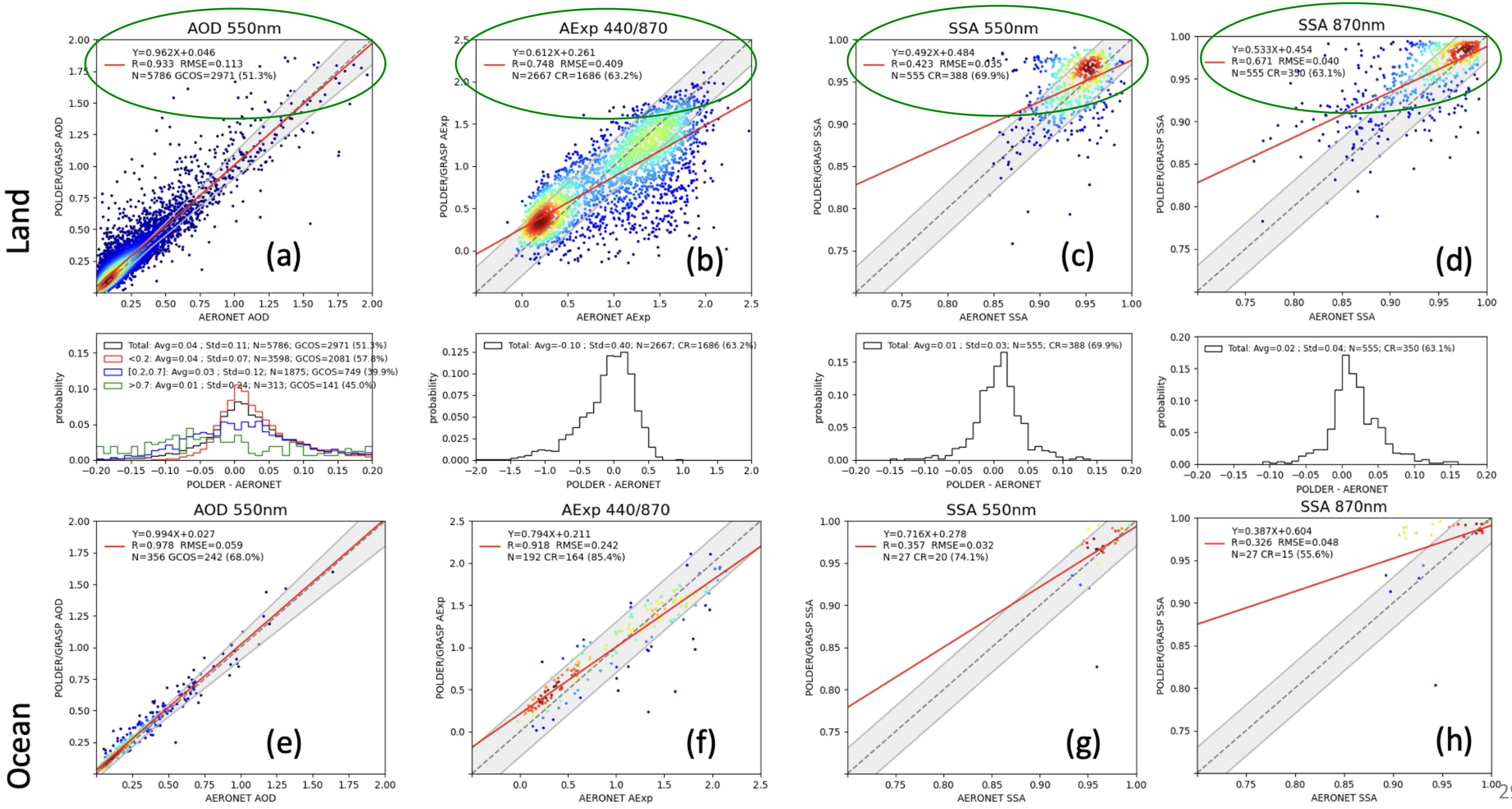
$$\hat{m}^{coarse_1}_{eff} = \hat{m}_{SeaS}\delta_{SeaS} + \hat{m}_{Water}\delta^{coarse}_{Water}$$

Coarse mode:

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

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

Improved performance



Current status of harmonization:

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

Current Questions:

CAMS / MERRA-2:

- assumptions of species ref. indices - ?;
(BC - ?)

- 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 modes 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.?)

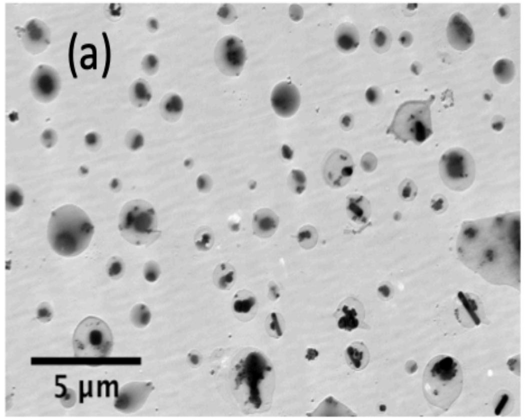
No degradation of real retrieval from modification!

Aerosol models in CAMS/MERRA-2 and remote sensing

	Aerosol modelling approach	CAMS/MERRA-2	Optimal model for multi-angular polarimetric remote sensing	Single/bi-viewing imagers	Lidars
1	**Aerosol modes and aerosol species**	• **5 aerosol species**, **7 components**: BC(2), OM(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	?	• ?

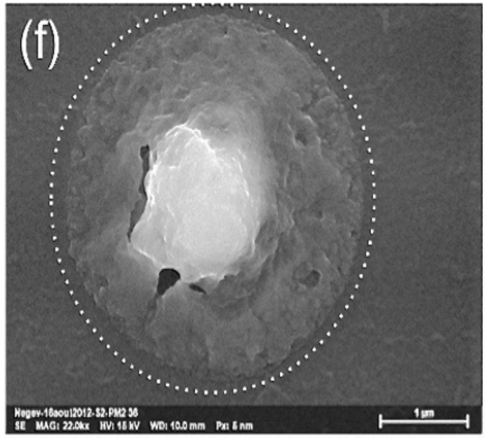
Aerosol non-sphericity and inhomogeneity <u>in remote sensing</u>

Urban aerosol (Lille, France)

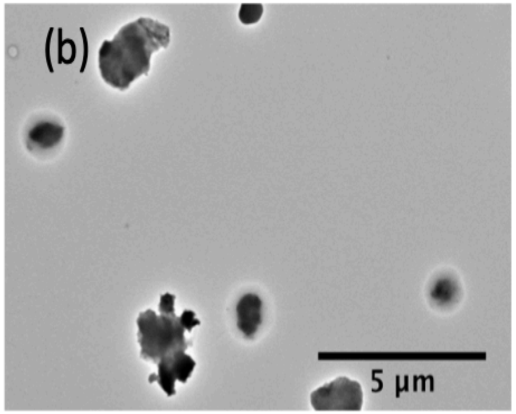


F. Unga, PhD thesis 2017, U. Lille

Desert aerosol (Senegal and Israel)

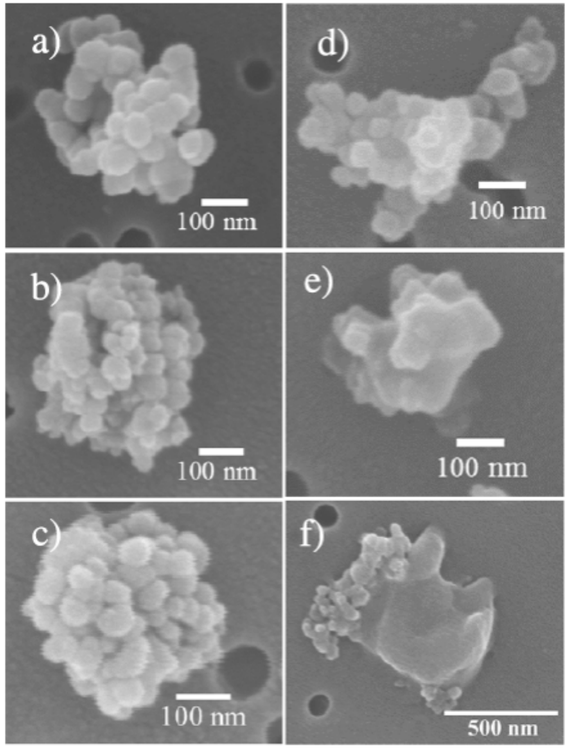


Derimian et al, ACP, 2017.

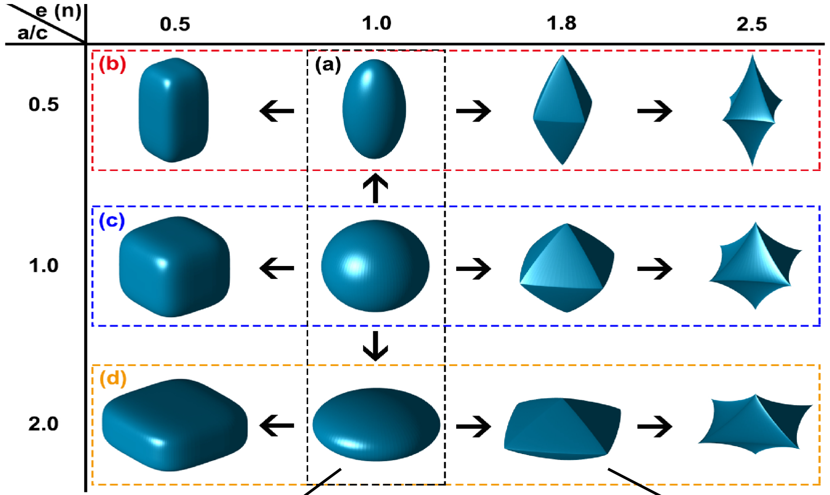


F. Unga, PhD thesis 2017, U. Lille
https://theses.fr/2017LIL10023

Coated soot

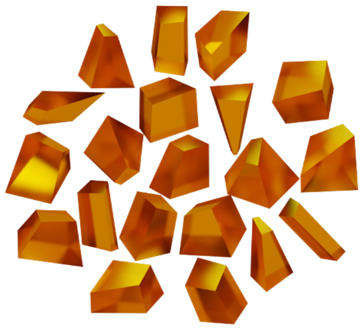


Model of fractal-like cluster of particles in the shell (under development in collaboration with V. Tyshkovets and L. Berdina)

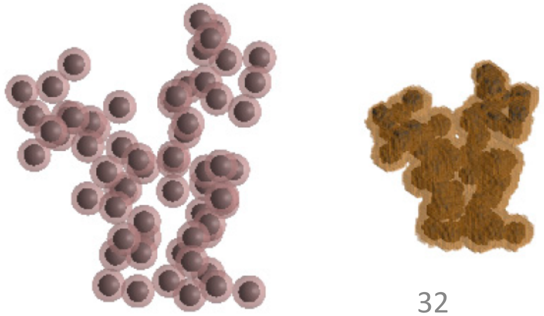


Spheroids
Dubovik et al., 2006

Super Spheroids
Bi et al., 2018

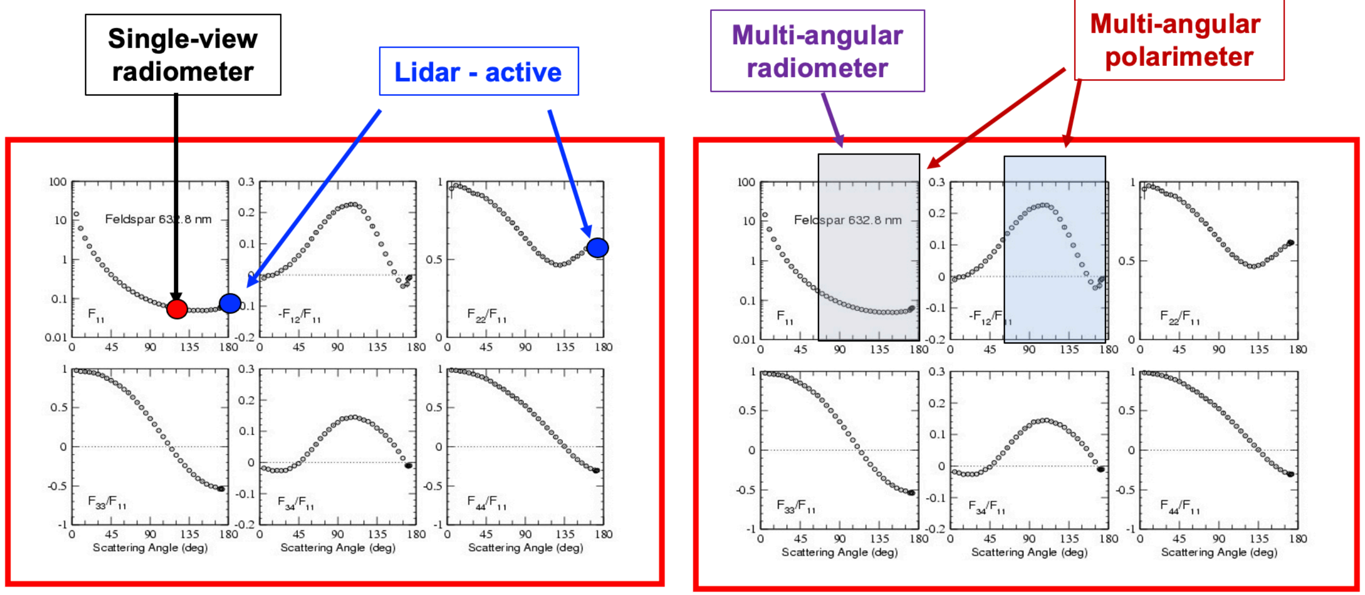


Hexahedrons
Saito et al., 2021 a,b

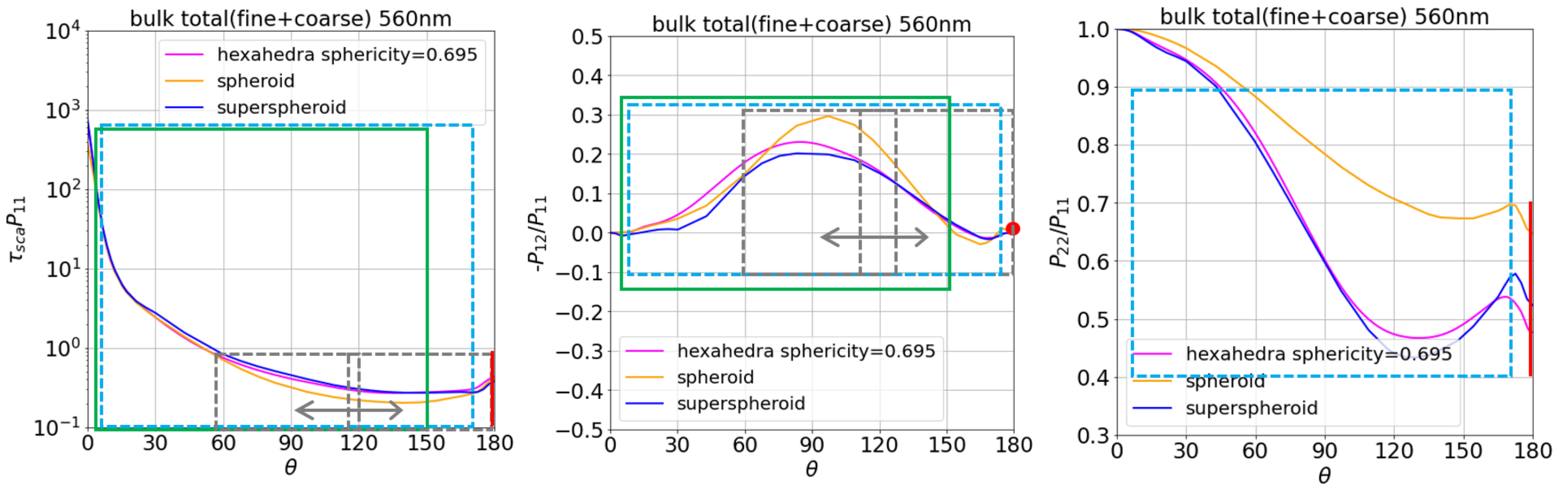


M. Kahnert, Opt. Express 2017

Particle shape effects on the observations



- Multi-wavelength lidar (~m^3)
- Spaceborne polarimeters (~km^3)
- Ground-based sun-photometers (~km^3)
- Polar nephelometers* (~dm^3)



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 MERA/CAMS?
   - ➢ Retrieved SD in remote sensing vs prescribed with accounting for hygroscopic growth in MERA/CAMS
5. Non-sphericity and inhomogeneity in MERA/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 MERA/CAMS?
   - ➢ AOD? What spectral bands? Fine mode AOD? Angstrom Exponent? SSA?

THANK YOU !