ICAP 9TH WORKING GROUP:
Radiative Transfer and Impacts of Aerosol Radiative Forcing on Numerical Weather Prediction

International Cooperative for Aerosol Predication
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CERLA, University of Lille, FR

Inferring emissions of desert dust and primary carbonaceous aerosol from PARASOL/GRASP retrievals

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June 28th , 2017

Why is aerosol emission?

Annual aerosol emission sources:

IPCC, 2013

Sulfate

87.8~167.5 Tg/yr

Sea Spray

1400~6800 Tg/yr

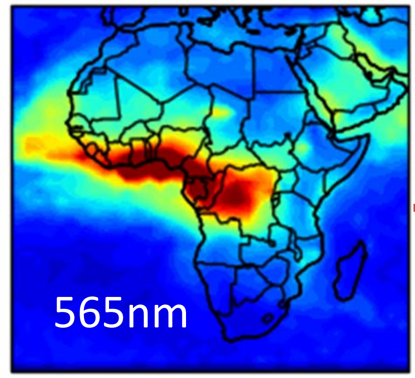
<u>High uncertainty in aerosol emission sources translates into a significant uncertainty in aerosol climate effects evaluation.</u>

Observation *vs.* Simulation

- Satellite Observations – PARASOL/GRASP
  - Products: spectral AOD & AAOD, AExp, SSA(λ)

EXAMPLES : PARASOL/GRASP

AOD 2008



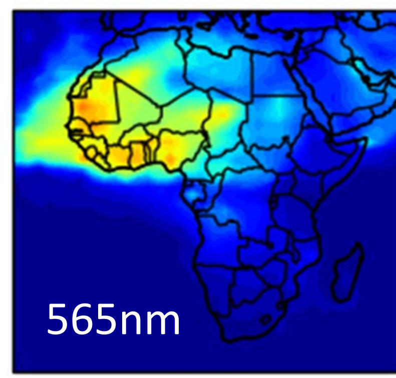
Difference?
Accuracy?

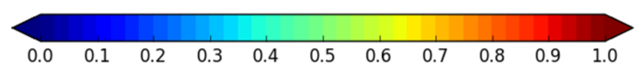
- Model Simulations – GEOS-Chem

Model emission sources
BC, OC, Dust, etc.

"Prior Emission Sources"

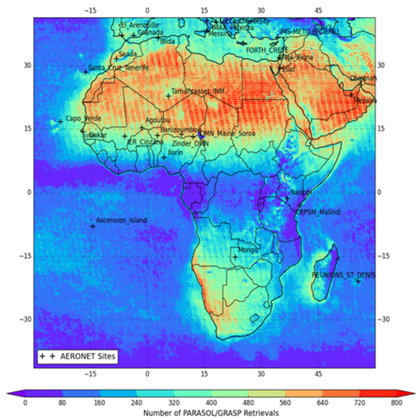
advection → diffusion → convection → deposition, etc.





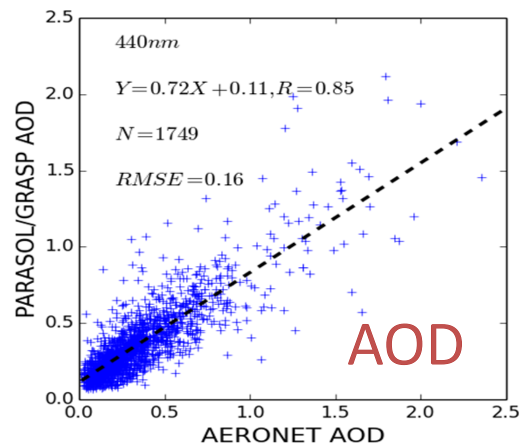
What's the accuracy?

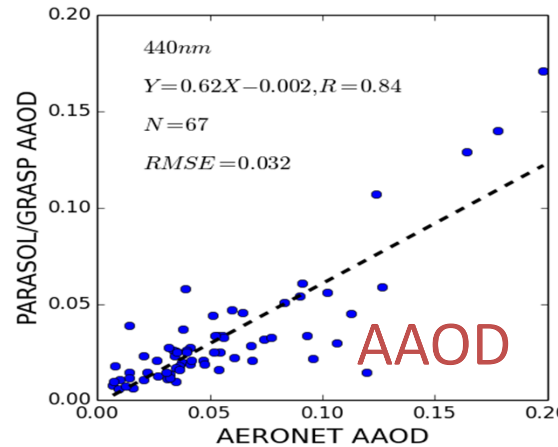
Validate over 28 AERONET sites



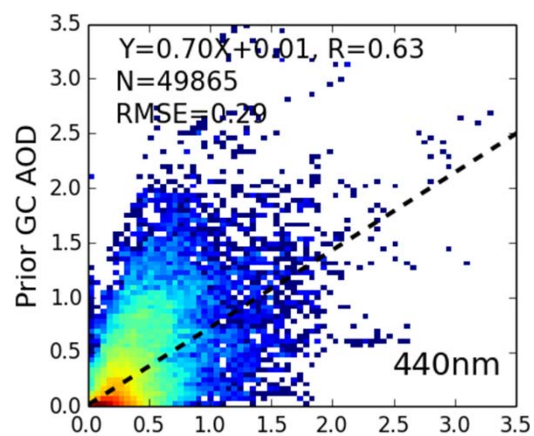
Re-grid
2° x2.5°

➢ PARASOL/GRASP *vs.* AERONET





➢ Prior GEOS-Chem *vs.* AERONET



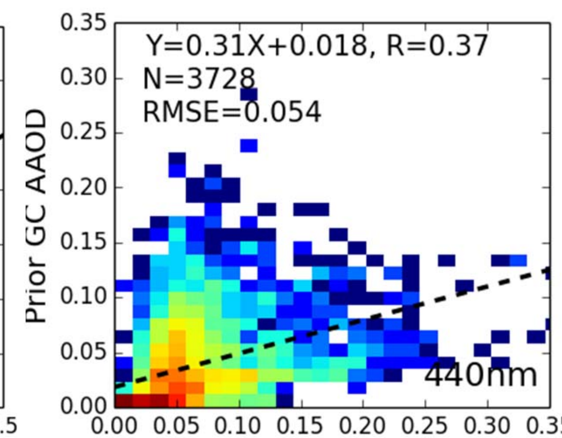


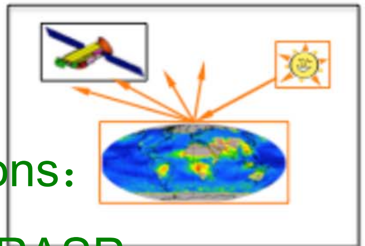
Table. Summary of Linear Regression

vs. AERONET	AOD	AAOD
PARASOL/GRASP	R=0.85; RMSE=0.16	R=0.84; RMSE=0.032
Prior GEOS-Chem	R=0.63; RMSE=0.29	R=0.37; RMSE=0.054

General Concept of Aerosol Emission Retrieval

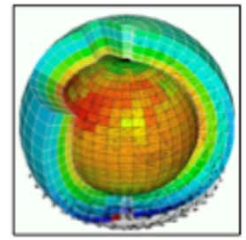
I. Observations：

PARASOL/GRASP



$$m(t,\boldsymbol{x}) = \int_{t_0}^{t} T(t',\boldsymbol{x})(m(t',\boldsymbol{x}) + s(t',\boldsymbol{x}))dt'$$

$$T(t,x) = T_q T_{q-1} \dots T_3 T_2 T_1$$



II. Model：

GEOS-Chem and its adjoint

Satellite data:
observations of ambient aerosol

INVERSION:
(e.g. via adjoint model)

Transport Model:
-meteorology
- transport
- chemistry
- removal
- emissions?

III. Inversion

Retrieved Emissions

"Posterior Emissions"

Improved sources (location and strength)
Improved agreement of modeling with observations
Improved aerosol characterization for climate, air quality,etc.

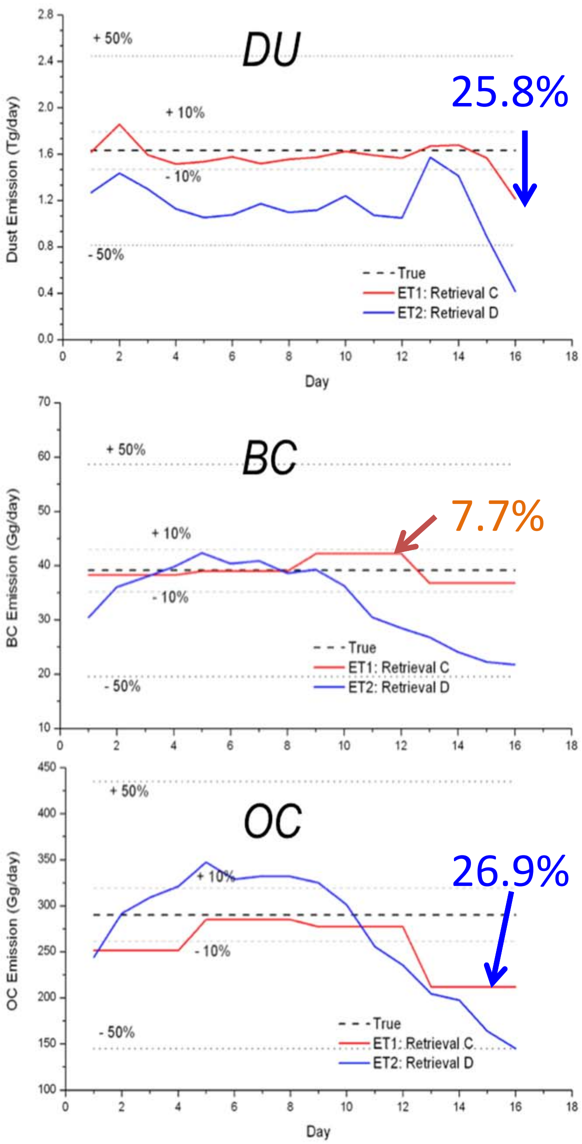
Cost Function: $\mathrm{J}(\boldsymbol{\sigma}) = \frac{1}{2} \sum_{\tau \in \tau, \tau_a} (\tau(\boldsymbol{\sigma}) - \tau_{obs})^{\mathrm{T}} \mathrm{C}_{obs}^{-1} (\tau(\boldsymbol{\sigma}) - \tau_{obs})$

Dubovik et al., ACP, 2008
Henze et al., ACP, 2007

Adjoint method: $\nabla_{\boldsymbol{\sigma}} \mathrm{J}(t, \boldsymbol{x}) = \int_{t}^{t_0} \mathrm{T}^{\mathrm{T}}(t', \boldsymbol{x})(\nabla(t', \boldsymbol{x}) + \Delta \mathrm{M}(t', \boldsymbol{x}))(-\mathrm{d}t')$

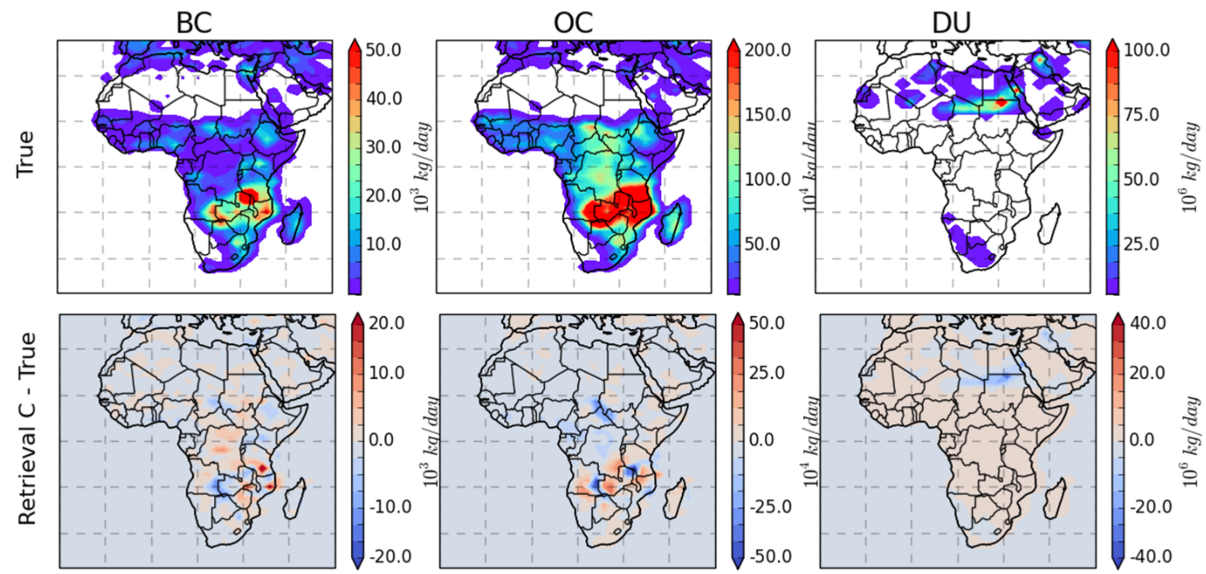
Spectral AOD and AAOD help to characterize DU, BC and OC aerosol emissions simultaneously.

Sensitivity Test – Invert synthetic measurements



Daily Total Emission

Figure. Spatial Distribution of true emission and the difference between retrieved and true emission



Summary:

- ✓ 6 wavelengths AOD and AAOD from PARASOL/GRASP input information are capable of determine the spatial distribution of BC, OC and DU emissions simultaneously.
- ✓ Uncertainty for daily total emission is ~26% for DU, ~8% for BC and ~ 27% for OC
- ✓ Uncertainty over the source region is less than ~30%
- ✓ The retrieval is initialized by “prior model” emissions with a background manually.
- ✓ The emissions are assumed daily constant for DU and 4 days constant for BC and OC.

Results – Invert PARASOL/GRASP real data

- Retrieved emission sources *vs.* model prior sources
- Spectral aerosol optical depth distribution simulated from retrieved sources *vs.* PARASOL / GRASP AOD
- Spectral absorption aerosol optical depth distribution simulated from retrieved sources *vs.* PARASOL / GRASP AAOD
- Comparison in statistics

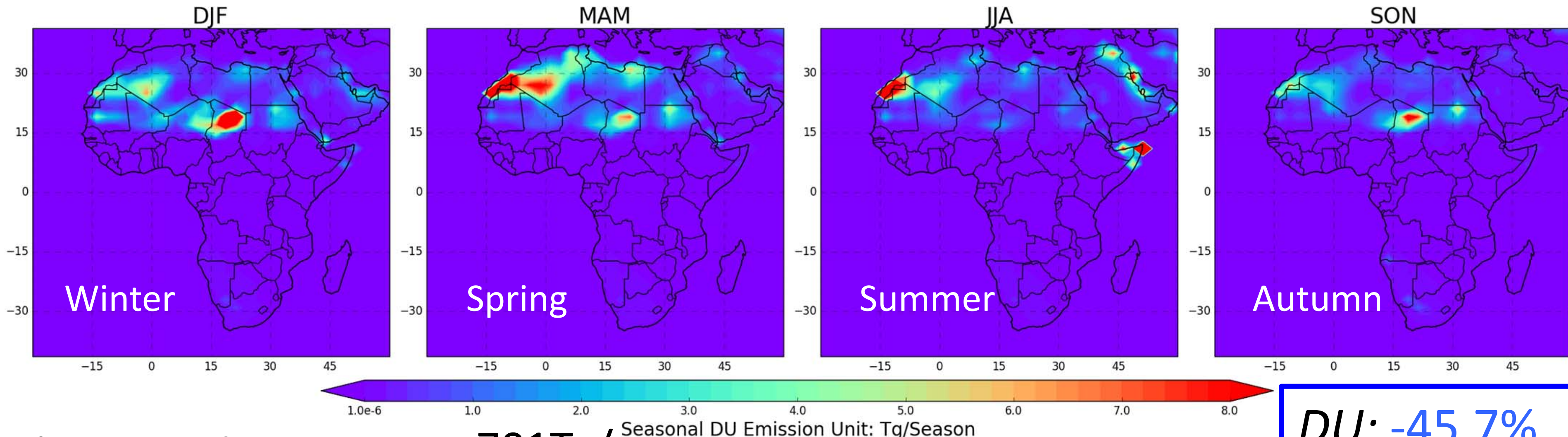
Study area: *Lat. [-40, 40], Lon. [-30, 60]*

Time period: December 2007 ~ November 2008

Retrieved vs. Model prior Dust emissions

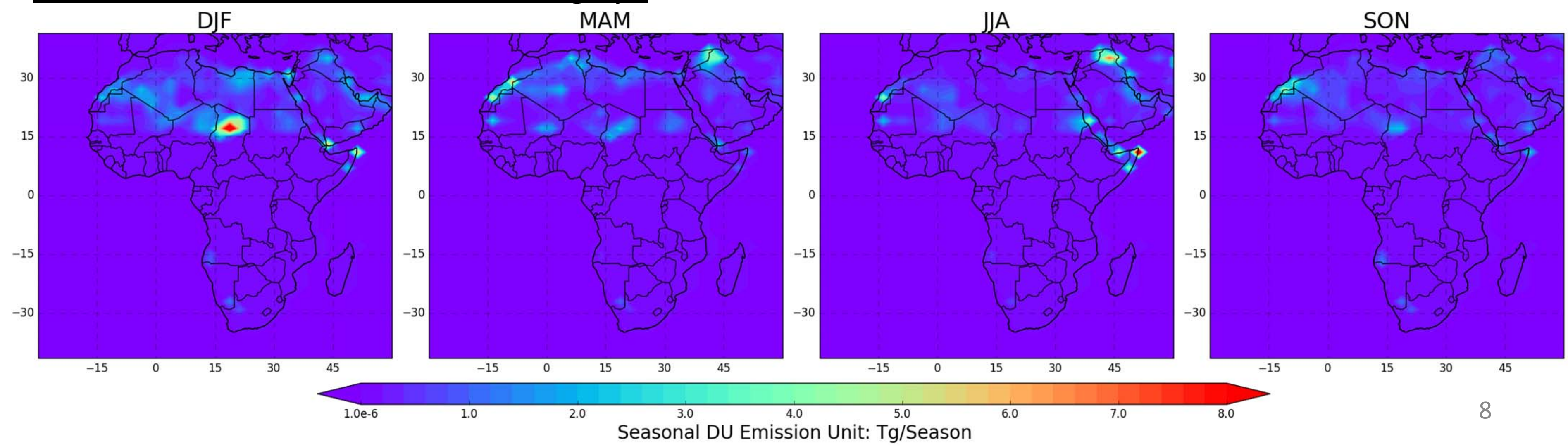
DEAD model (Zender et al., 2003)

a. Prior DU Emissions: 1291Tg/yr



DU: -45.7%

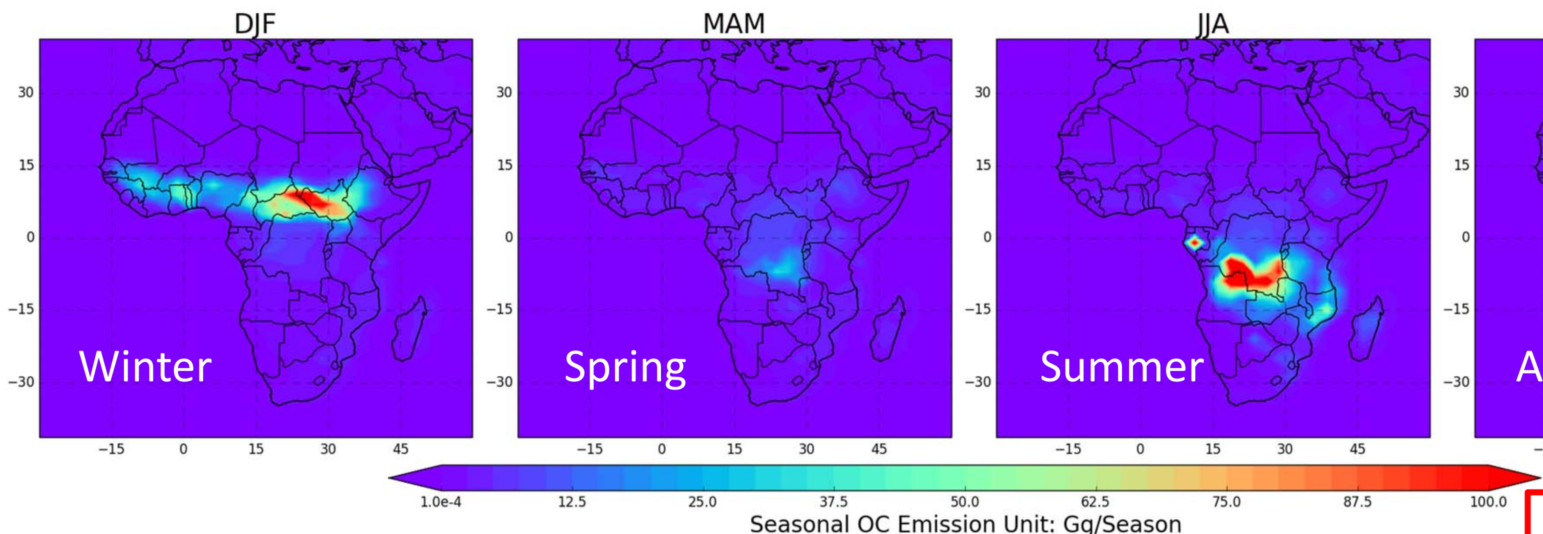
b. Retrieved DU Emissions: 701Tg/yr



Retrieved vs. Model prior Organic Carbon emissions

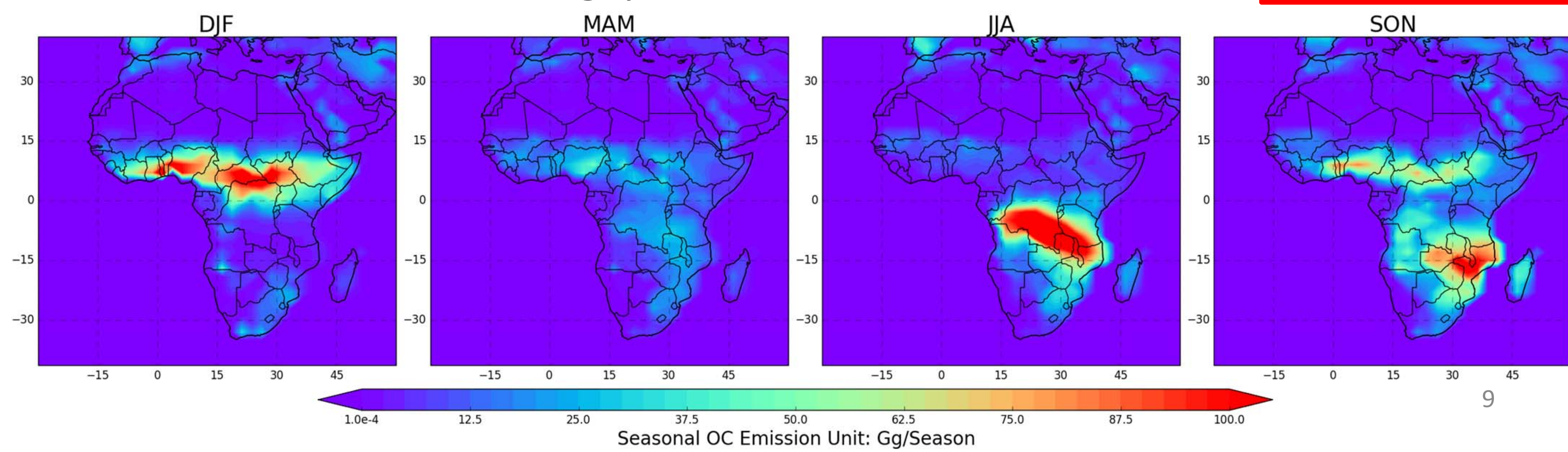
"Prior OC": Bond anthropogenic inventory and GFED3 inventory

a. Prior OC Emissions: 12.5Tg/yr



b. Retrieved OC Emissions: 37.1Tg/yr

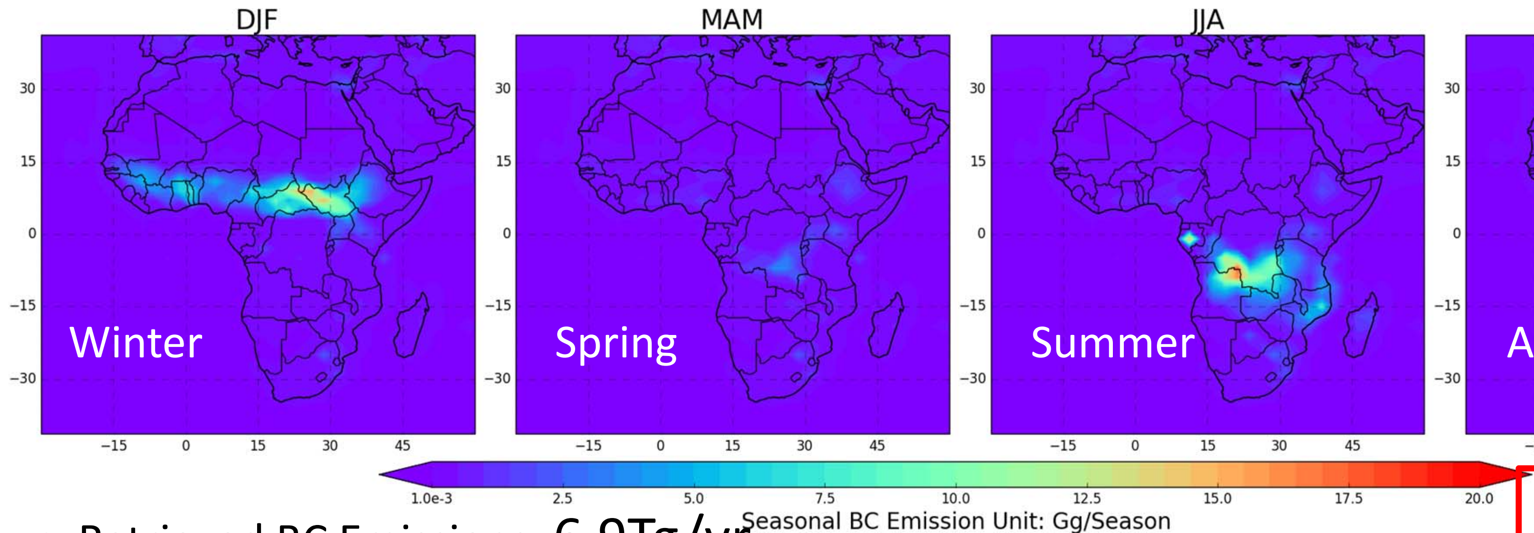
OC: +196.8%



Retrieved vs. Model prior Black Carbon emissions

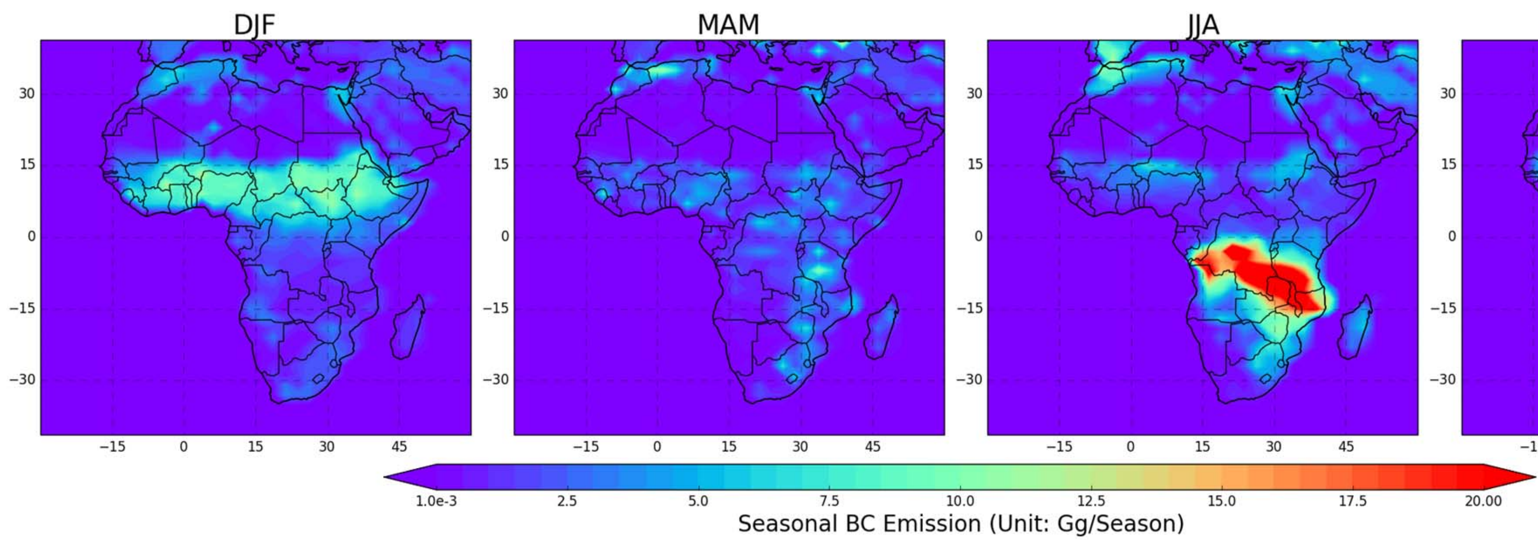
"Prior BC": Bond anthropogenic inventory and GFED3 inventory

a. Prior BC Emissions: 1.7Tg/yr

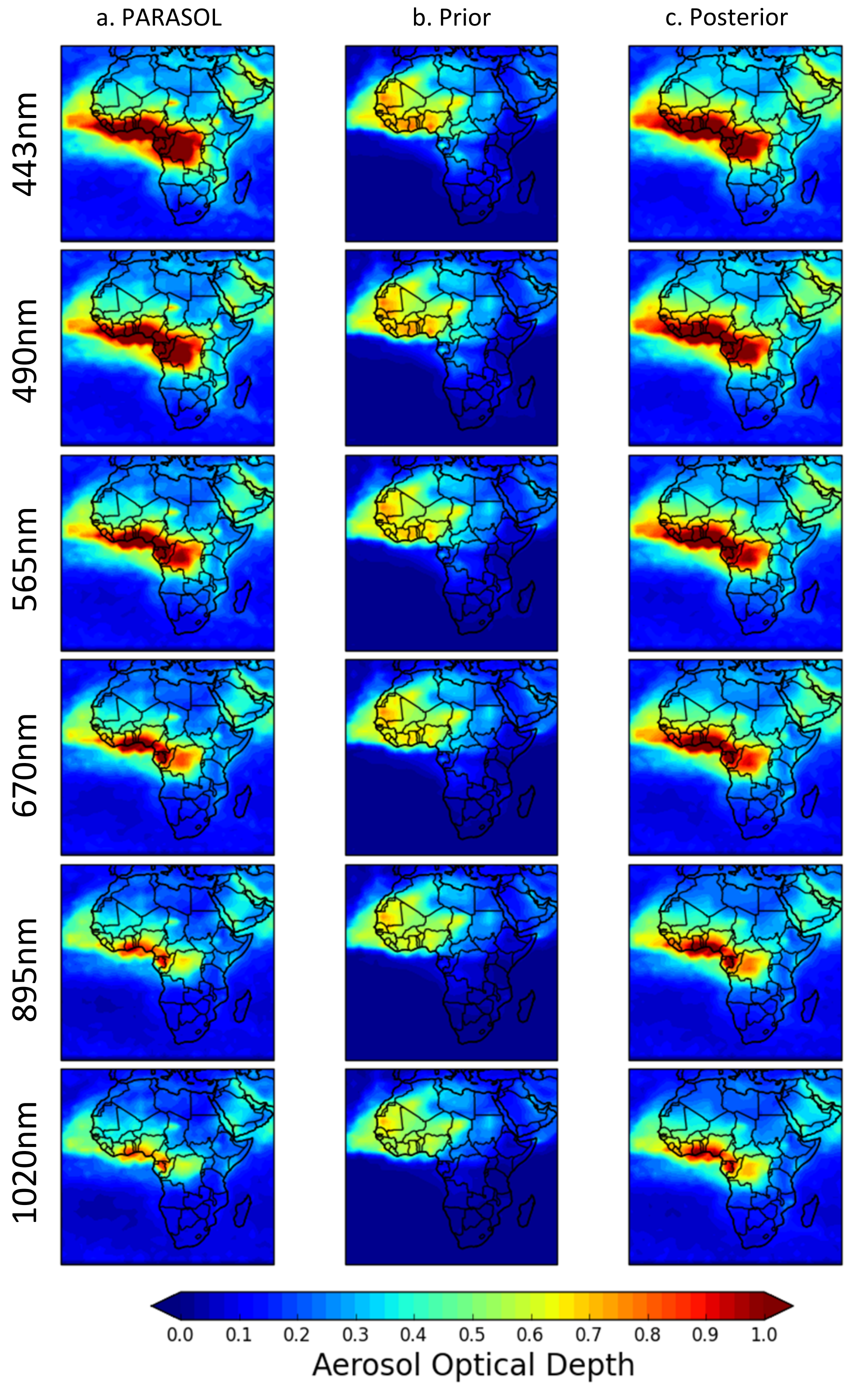


BC: +305.9%

c. Retrieved BC Emissions: 6.9Tg/yr



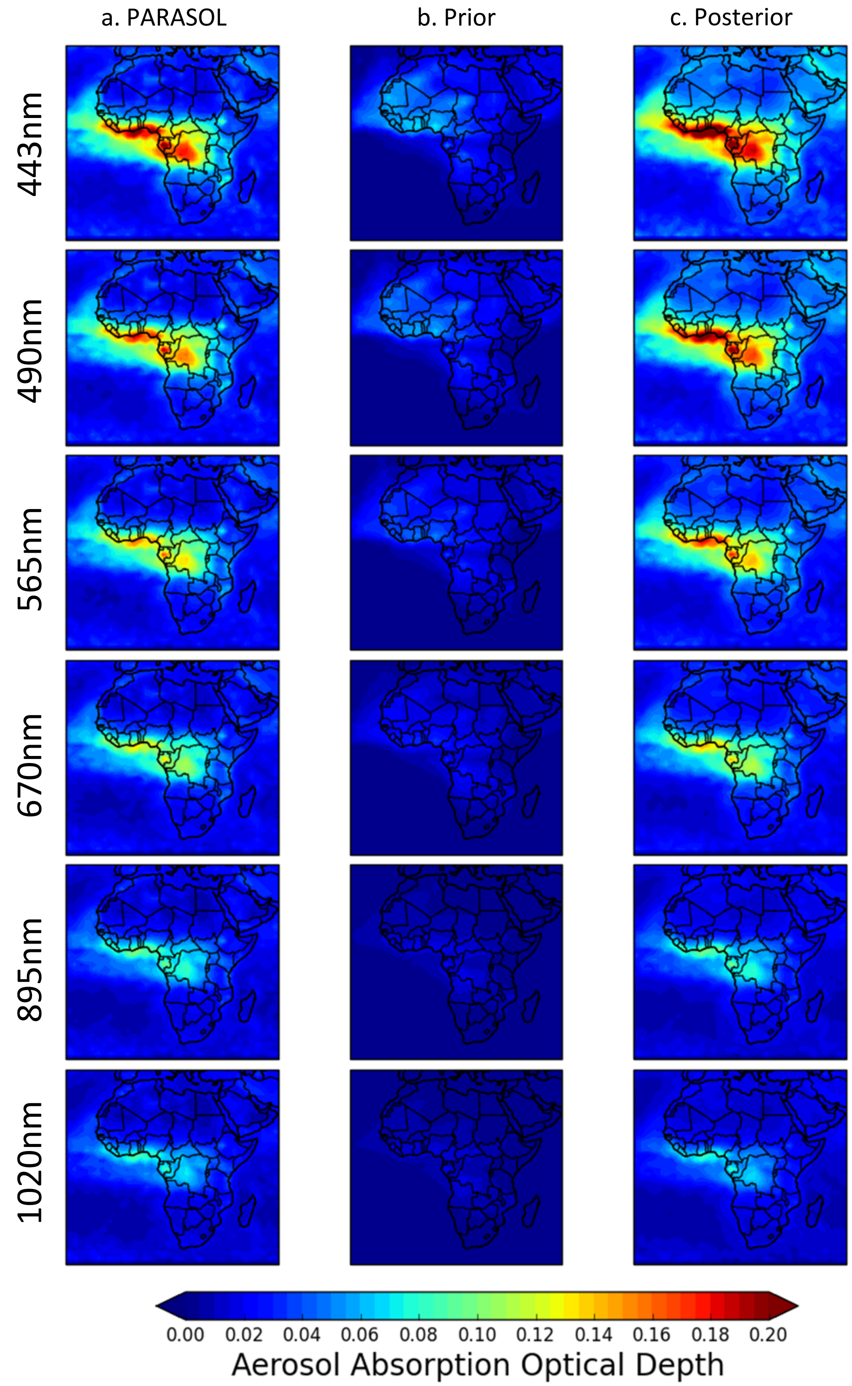
Fit of Aerosol Optical Depth



a. PARASOL/GRASP AOD
b. GEOS-Chem simulated AOD with prior model aerosol emission
c. GEOS-Chem simulated AOD with retrieved DU, BC and OC aerosol emission

Table. Summary of Linear Regression

	Prior vs. PARASOL	Posterior vs. PARASOL
443	R=0.48; E=0.34	R=0.92; E=0.13
490	R=0.48; E=0.33	R=0.92; E=0.12
565	R=0.49; E=0.31	R=0.92; E=0.12
670	R=0.50; E=0.30	R=0.92; E=0.12
865	R=0.51; E=0.28	R=0.91; E=0.11
1020	R=0.51; E=0.27	R=0.89; E=0.10



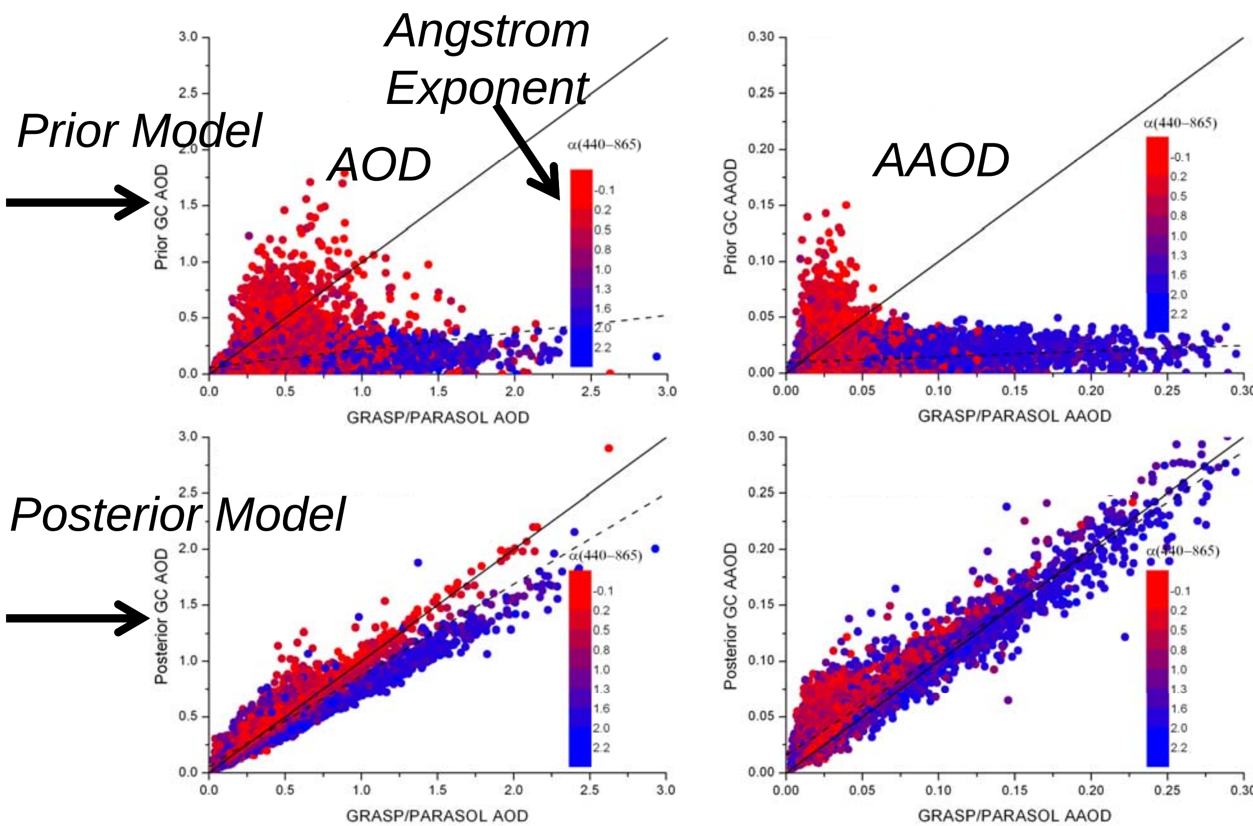
Fit of Aerosol Absorption Optical Depth

a. PARASOL/GRASP AAOD
b. GEOS-Chem simulated AAOD with prior model aerosol emission
c. GEOS-Chem simulated AAOD with retrieved DU, BC and OC aerosol emission

Table. Summary of Linear Regression

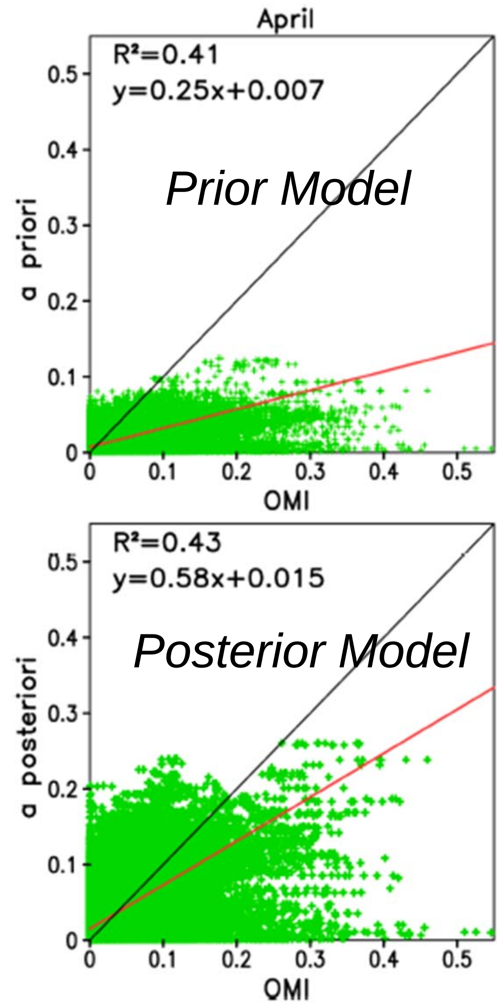
	Prior vs. PARASOL	**Posterior vs. PARASOL**
443	R=0.14; E=0.044	R=0.91; E=0.023
490	R=0.14; E=0.039	R=0.91; E=0.019
565	R=0.14; E=0.034	R=0.92; E=0.015
670	R=0.20; E=0.029	R=0.92; E=0.010
865	R=0.33; E=0.025	R=0.91; E=0.008
1020	R=0.30; E=0.022	R=0.90; E=0.008

Prior/Posterior Model vs. PARASOL/GRASP AOD and AAOD



Compare with Other studies

Zhang et al., ACP, 2015

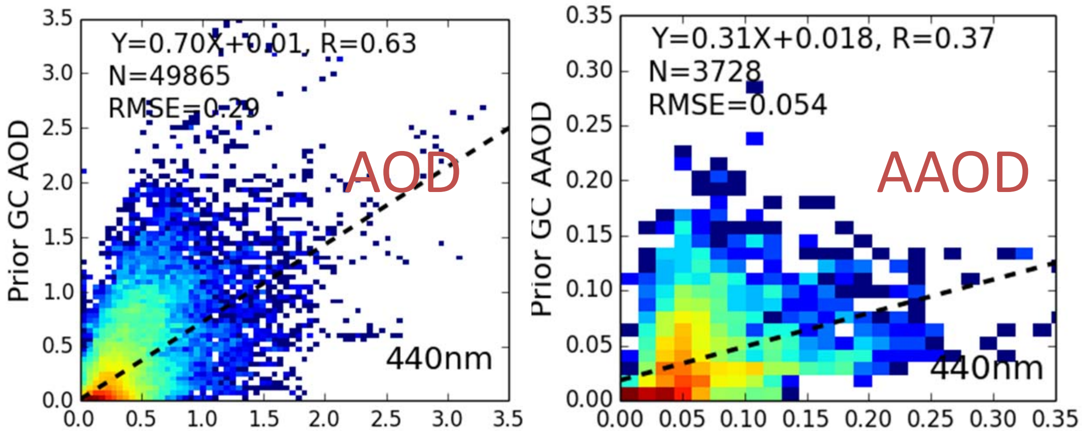


Reasons for better fit of observations:

- ✓ PARASOL/GRASP spectral AOD and AAOD allow us to determine DU, BC and OC emission simultaneously, correcting the major absorbers together is easier to find the best fit.
- ✓ In retrieval, we weak the constrain of prior knowledge of emission locations, which provide a better fit than constrain in particular pixels.

Validation with Independent AERONET Measurements

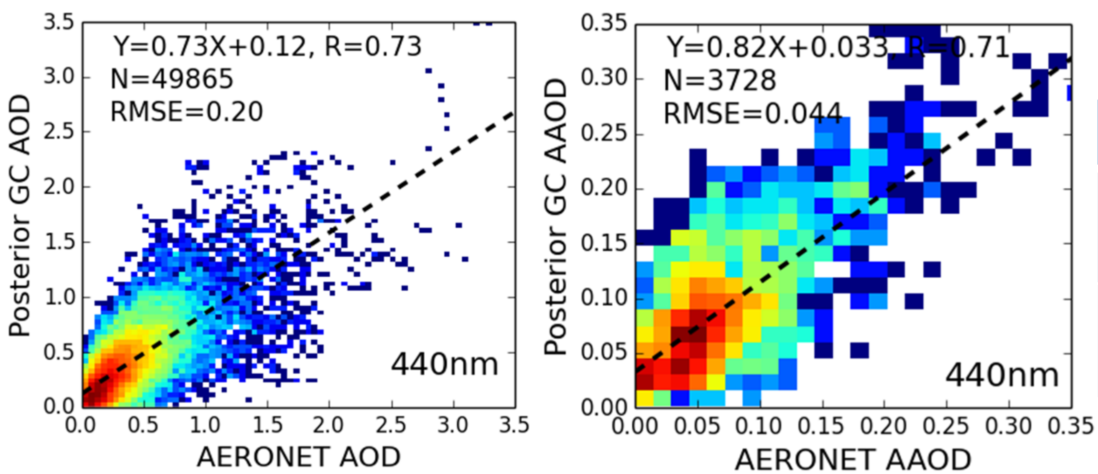
- Prior GEOS-Chem *vs.* AERONET



AERONET independent measurements: We didn't use AERONET data in retrieval.

- Posterior GEOS-Chem *vs.* AERONET

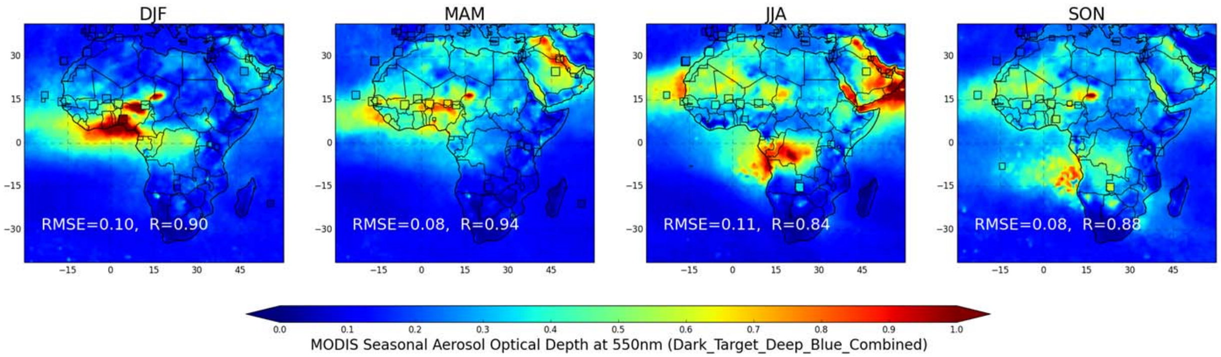
Posterior GEOS-Chem: Simulation with retrieved DU, BC and OC aerosol emissions.



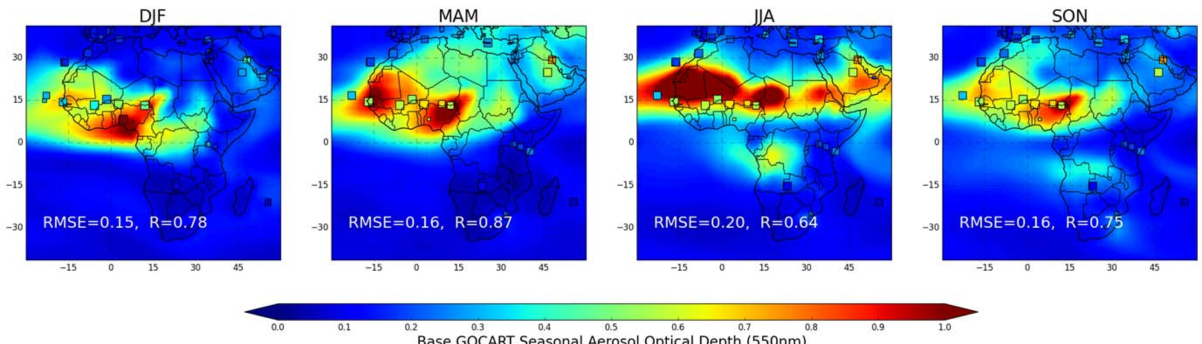
vs. AERONET	AOD	AAOD
Prior GEOS-Chem	R=0.63; RMSE=0.29	R=0.37; RMSE=0.054
Posterior GEOS-Chem	R=0.73; RMSE=0.20	R=0.71; RMSE=0.044

Implement retrieved emission into GEOS-5/GOCART model

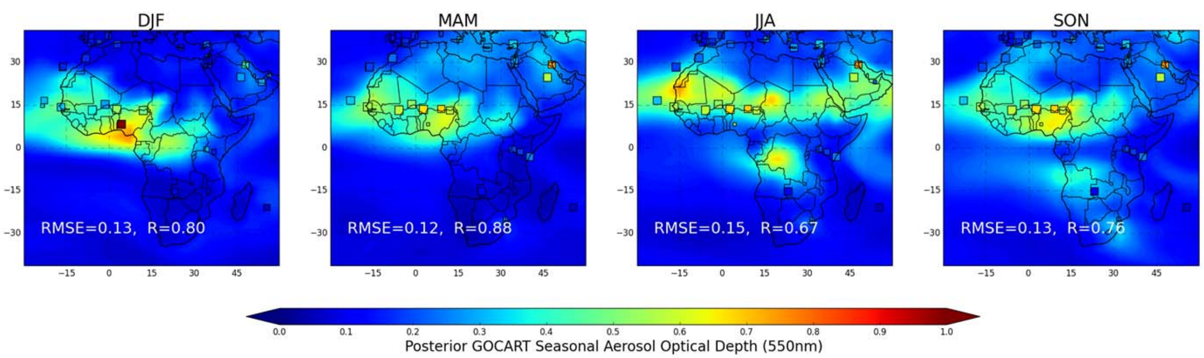
a. MODIS Seasonal AOD at 550nm

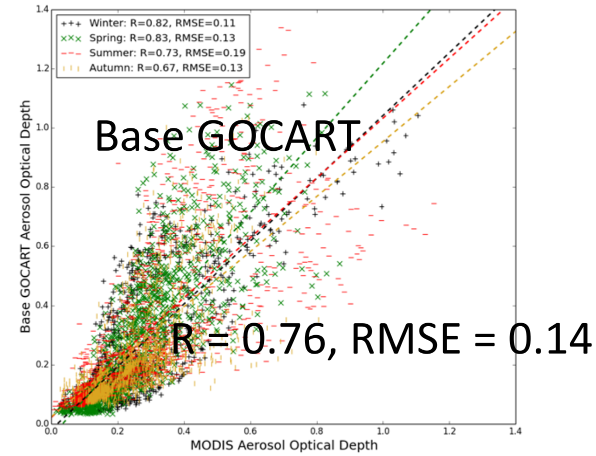


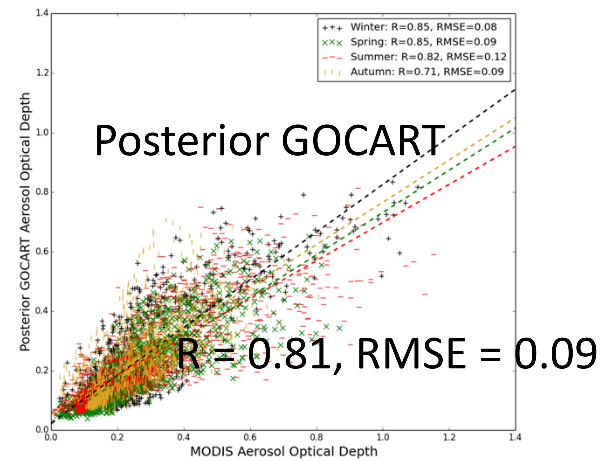
b. GEOS-5/GOCART base Seasonal AOD at 550nm



c. GEOS-5/GOCART posterior Seasonal AOD at 550nm



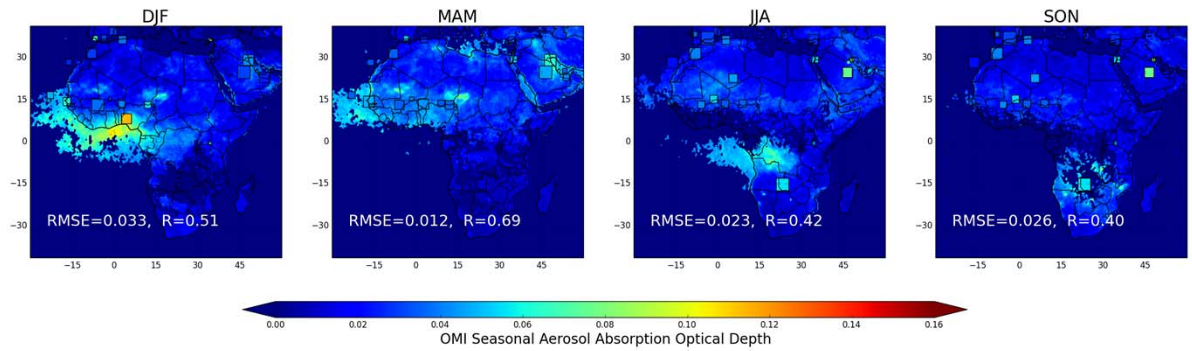




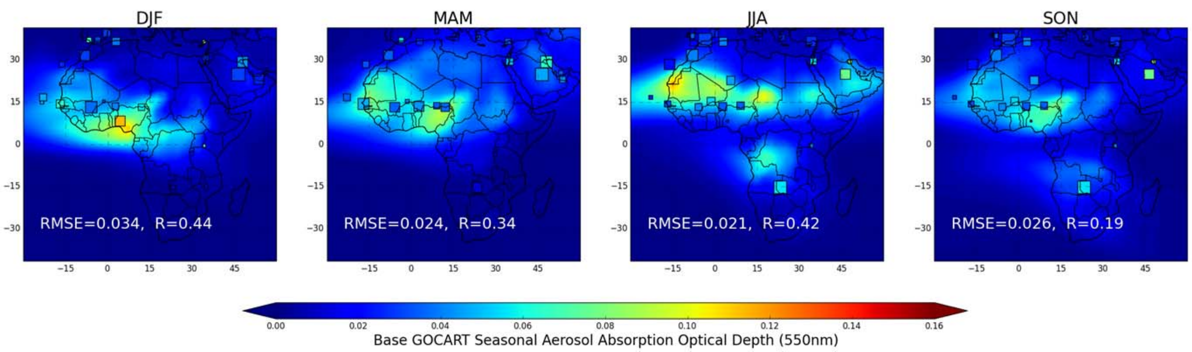
Note: GOCART AOD is daily 24h average, MODIS overpassing time is at noon.

AAOD

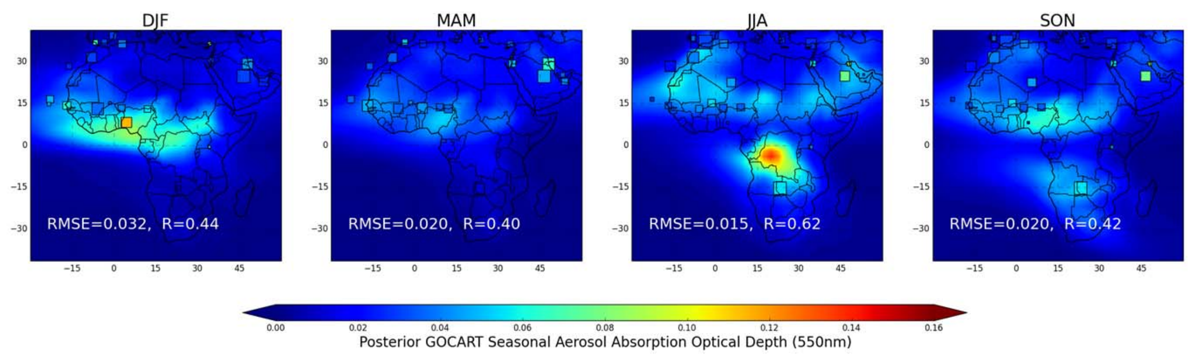
a. OMI Seasonal AAOD at 500nm



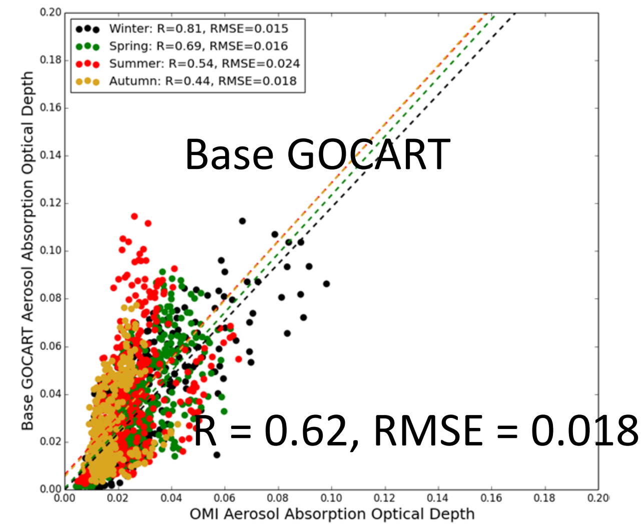
b. GEOS-5/GOCART base Seasonal AAOD at 550nm



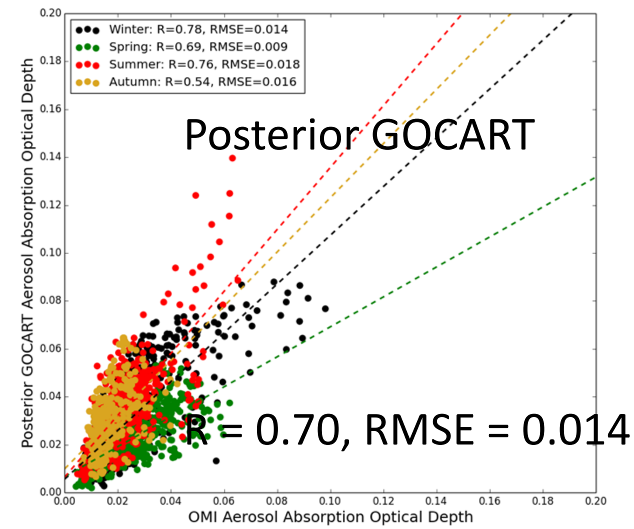
c. GEOS-5/GOCART posterior Seasonal AAOD at 550nm



d. GEOS-5/GOCART base *vs.* OMI



e. GEOS-5/GOCART posterior *vs.* OMI



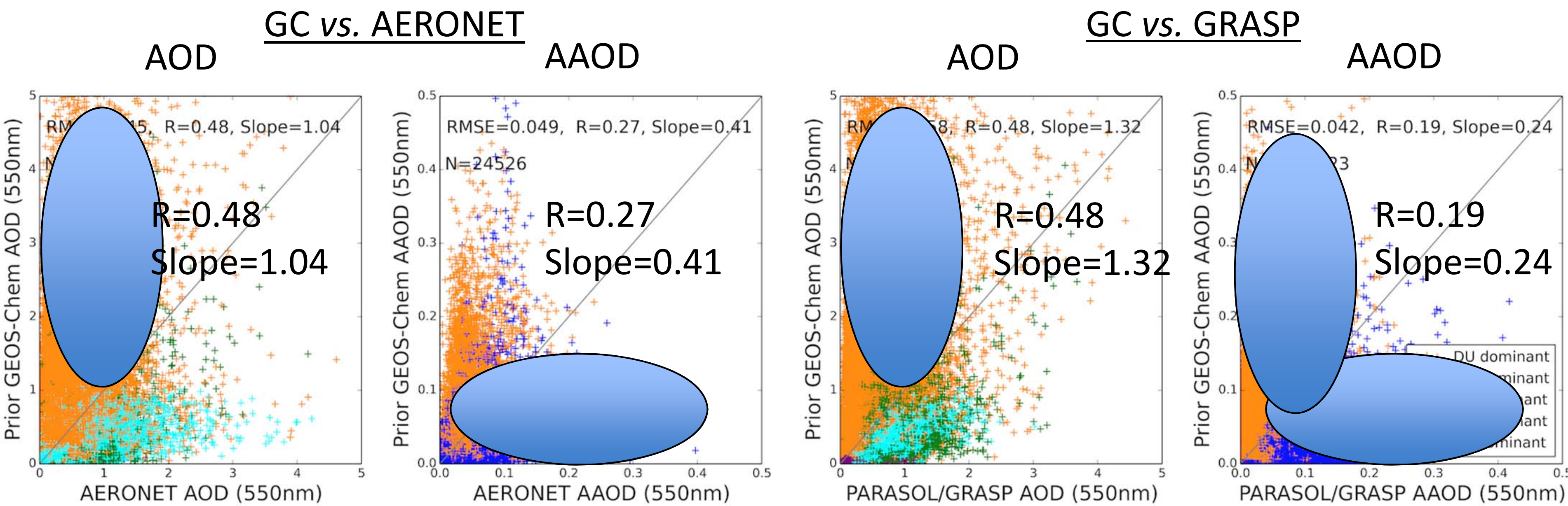
The significant increment of correlation coefficient shows in summer and autumn, which suggests more reliability of posterior aerosol emission at high biomass burning aerosol loading seasons.

Preliminary global results

A satellite view of global desert dust and primary carbonaceous aerosol emission database, 2006-2011

- ✓ Model evaluation with AERONET and Satellite observations
- ✓ Description of satellite based DU, BC and OC emission databases
- ✓ Posterior model simulation with new emission databases
- ✓ Perspectives and conclusions

Prior GEOS-Chem evaluation with AERONET and PARASOL/GRASP observations



AOD dominant: max{AOD_{BC}/AOD, AOD_{OC}/AOD, AOD_{DU}/AOD, AOD_{SU}/AOD, AOD_{SS}/AOD }
AAOD dominant: max{$AAOD_{BC}$/AAOD, $AAOD_{OC}$/AAOD, $AAOD_{DU}$/AAOD, $AAOD_{SU}$/AAOD, $AAOD_{SS}$/AAOD }

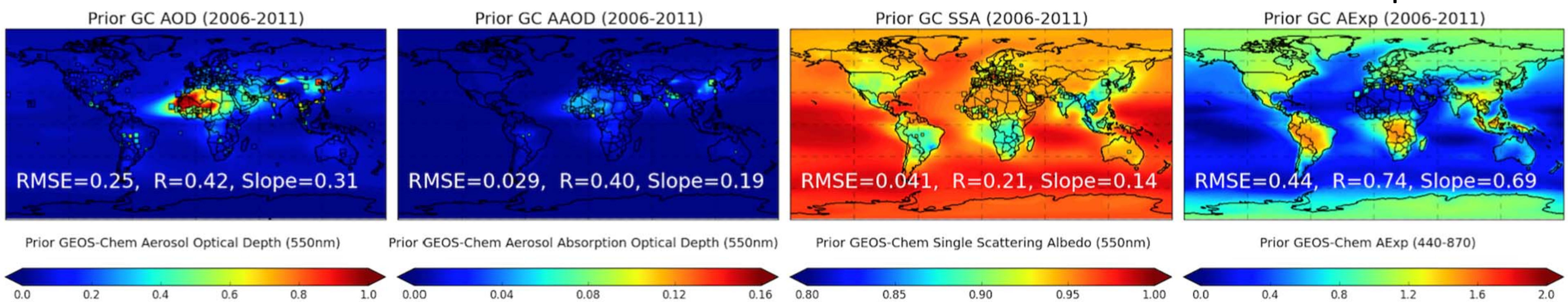
Major findings:

1. Dust AOD and AAOD from the model are broadly overestimated.
2. BC and dust are the two major components dominate the AAOD. However, the BC AAOD is underestimated.
3. The prior model AAOD is significant underestimated, with the linear regression slope ~0.41 with AERONET and ~0.24 with PARASOL/GRASP.

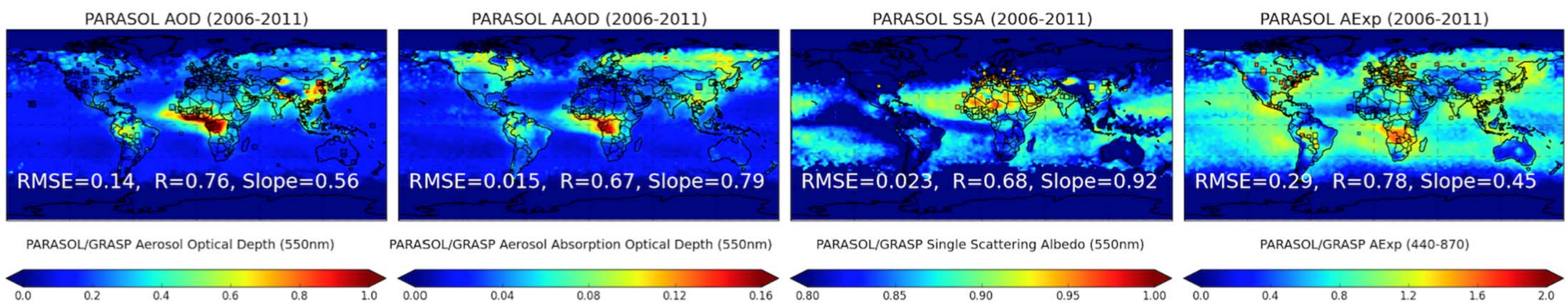
Prior AOD, AAOD, SSA and AExp (2006-2011)

Prior GEOS-Chem *vs.* AERONET

AOD | AAOD | SSA | AExp



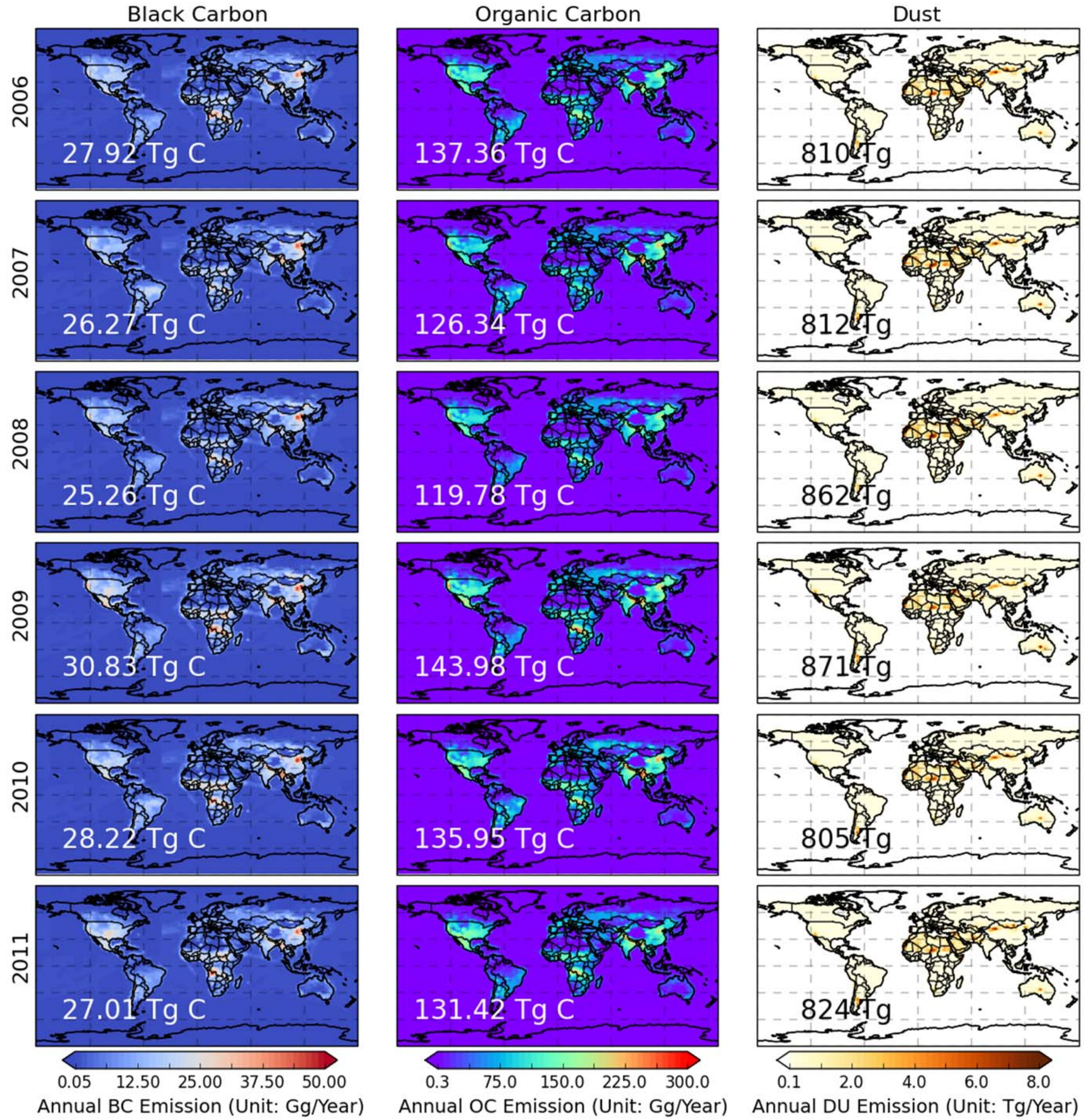
PARASOL/GRASP *vs.* AERONET



462 Sites

	AOD	AAOD	SSA	AExp
GEOS-Chem	R=0.42; RMSE=0.25	R=0.40; RMSE=0.029	R=0.21; RMSE=0.041	R=0.74; RMSE=0.44
GRASP 2° x2.5°	R=0.76; RMSE=0.14	R=0.67; RMSE=0.015	R=0.68; RMSE=0.023	R=0.78; RMSE=0.029

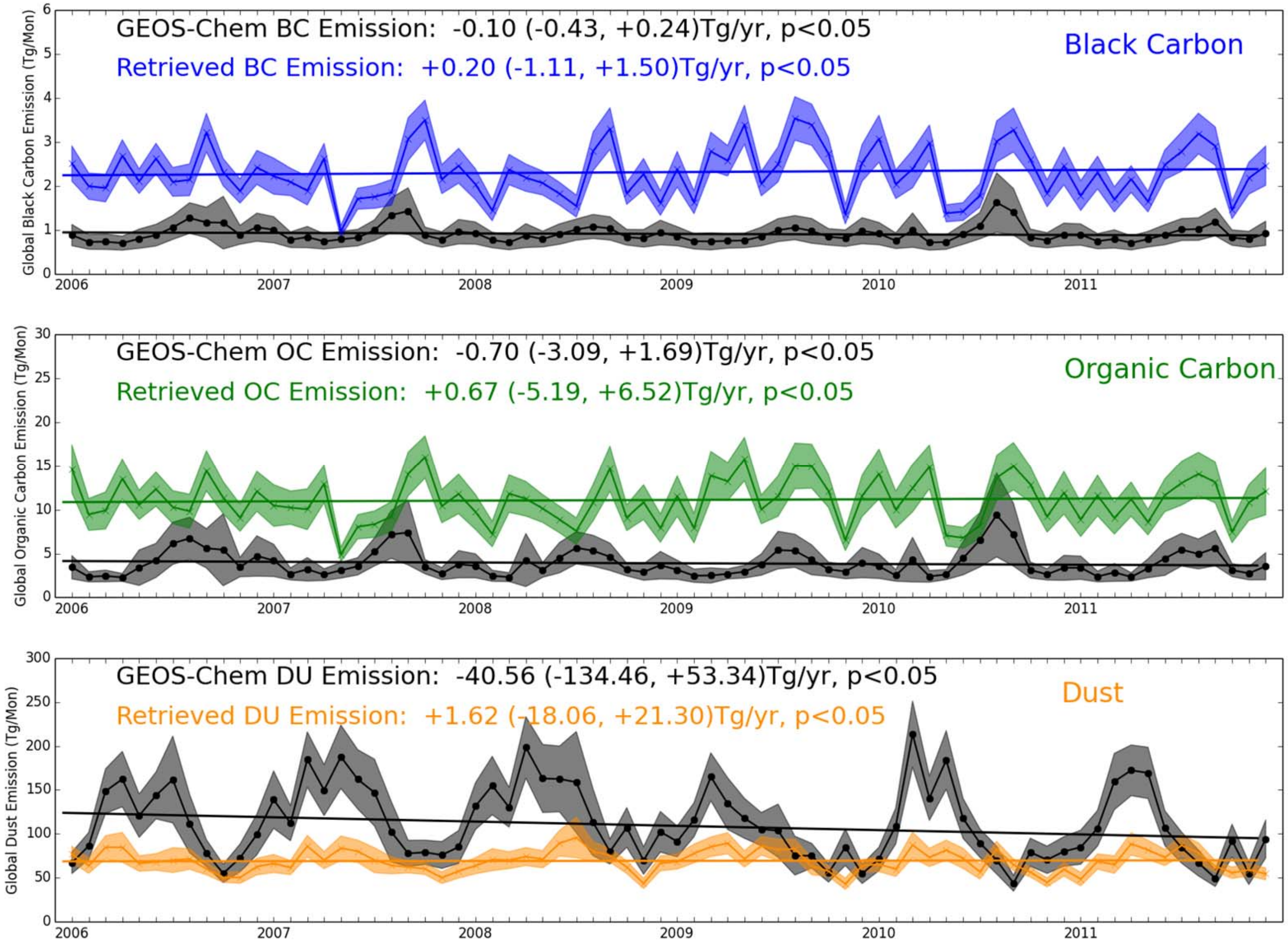
GRASP-based aerosol emission database (2006-2011)



GRASP-based aerosol emission database from 2006-2011.
Spatial resolution: 2° x2.5°
Emission Time resolution:
DU – 24 hours constant
BC – 48 hours constant
OC – 48 hours constant
Dust: 0.1 ~ 4.5 um (exclude super coarse particles)

Annual Mean (2006-2011)
BC: 27.6 Tg/yr
OC: 132.5 Tg/yr
DU: 831 Tg/yr

Comparison with prior GEOS-Chem inventories



BC: +152%

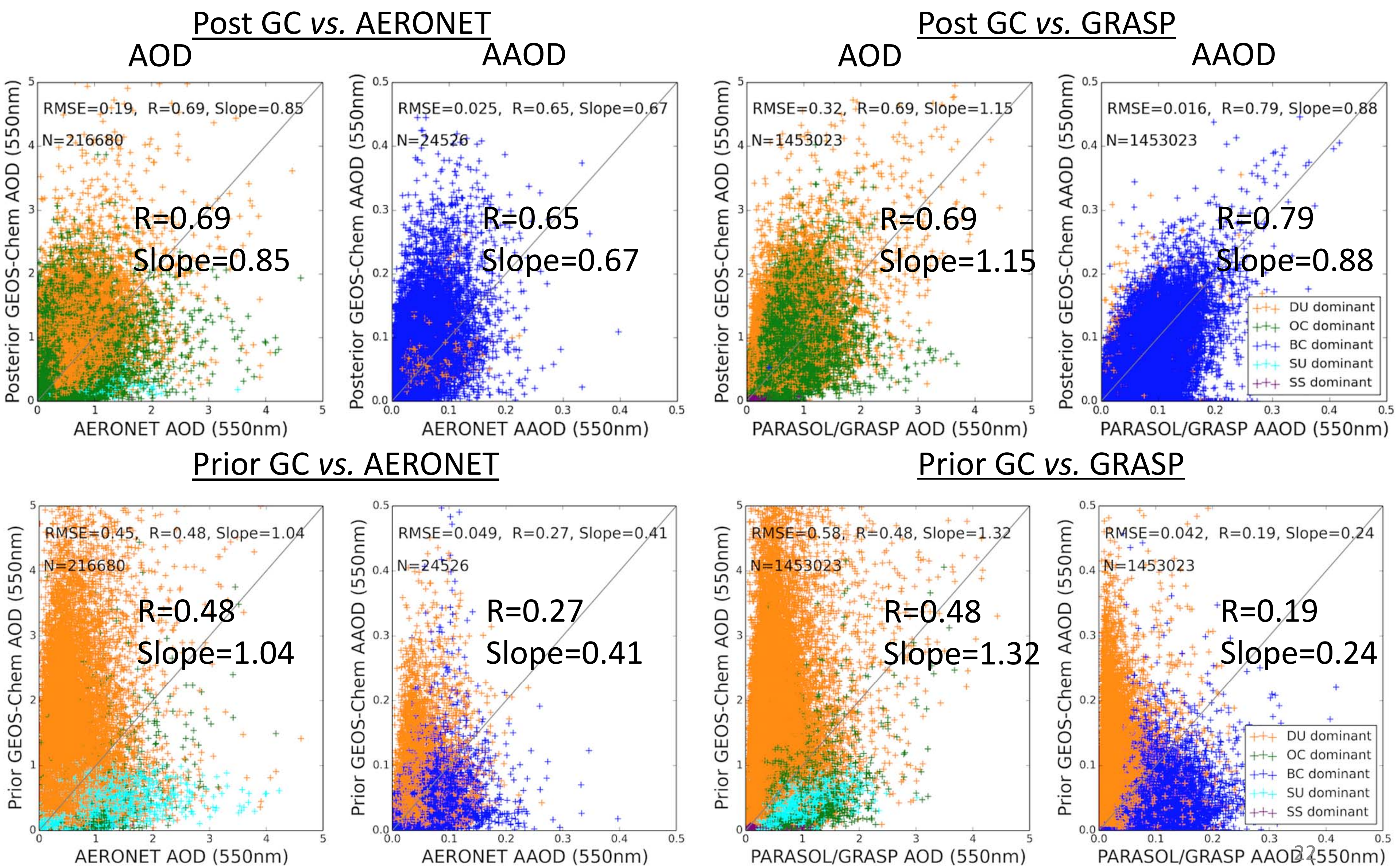
OC: +181%

DU: -38%

Dust: DEAD dust model (Zender et al., 2003)

Carbonaceous: Bond anthropogenic inventory + GFED3 BB inventory

Posterior *vs.* Prior GEOS-Chem simulation

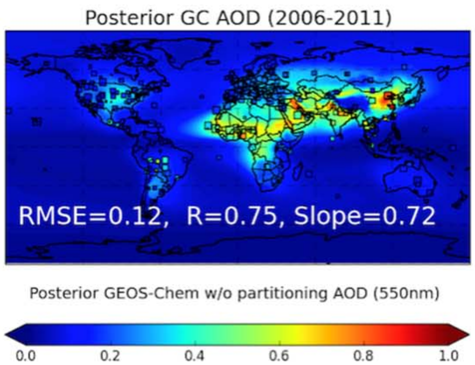


Posterior AOD, AAOD, SSA and AExp (2006-2011)

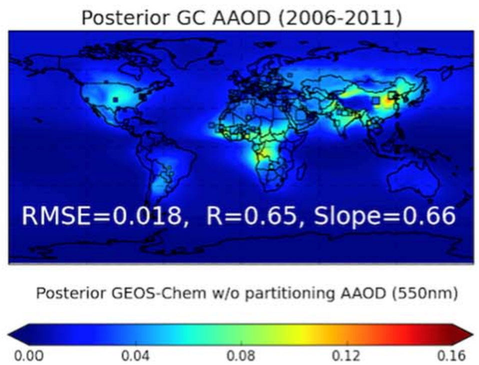
Annual Mean (2006-2011)

Posterior GEOS-Chem *vs.* AERONET

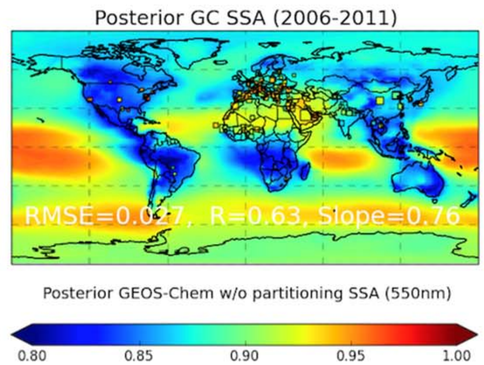
AOD



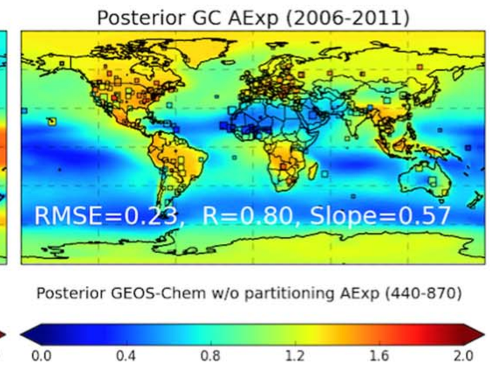
AAOD



SSA



AExp

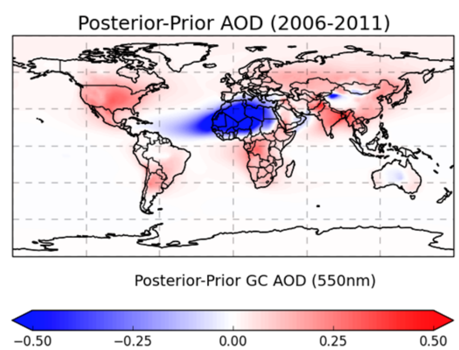


GEOS-Chem *vs.* AERONET

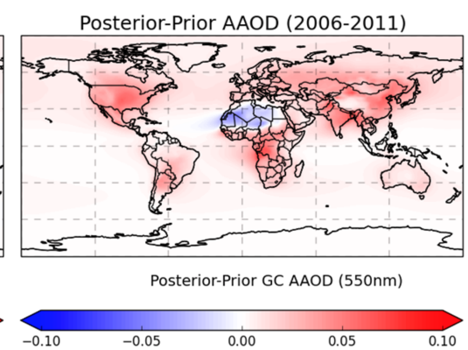
	Prior	Posterior
AOD	R=0.42; RMSE=0.25	R=0.75; RMSE=0.12
AAOD	R=0.40; RMSE=0.029	R=0.65; RMSE=0.018
SSA	R=0.21; RMSE=0.041	R=0.63; RMSE=0.022
AExp	R=0.74; RMSE=0.44	R=0.80; RMSE=0.23

Posterior – Prior

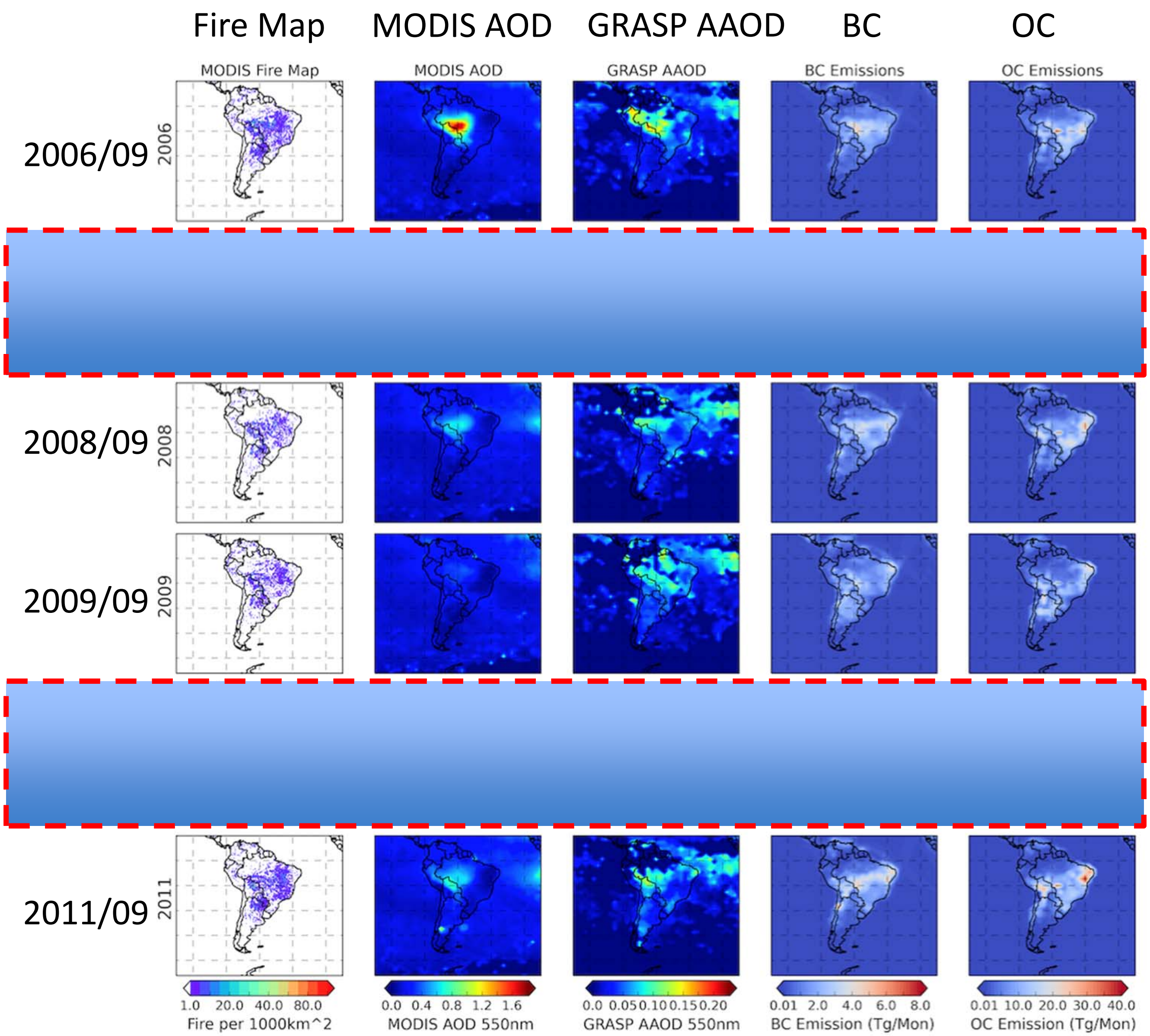
Diff. AOD



Diff. AAOD



Case Study: Annual cycle of South America biomass burning



September

Determine aerosol from satellite observations help to capture the inter-annual emission intensity and the regional extreme events.

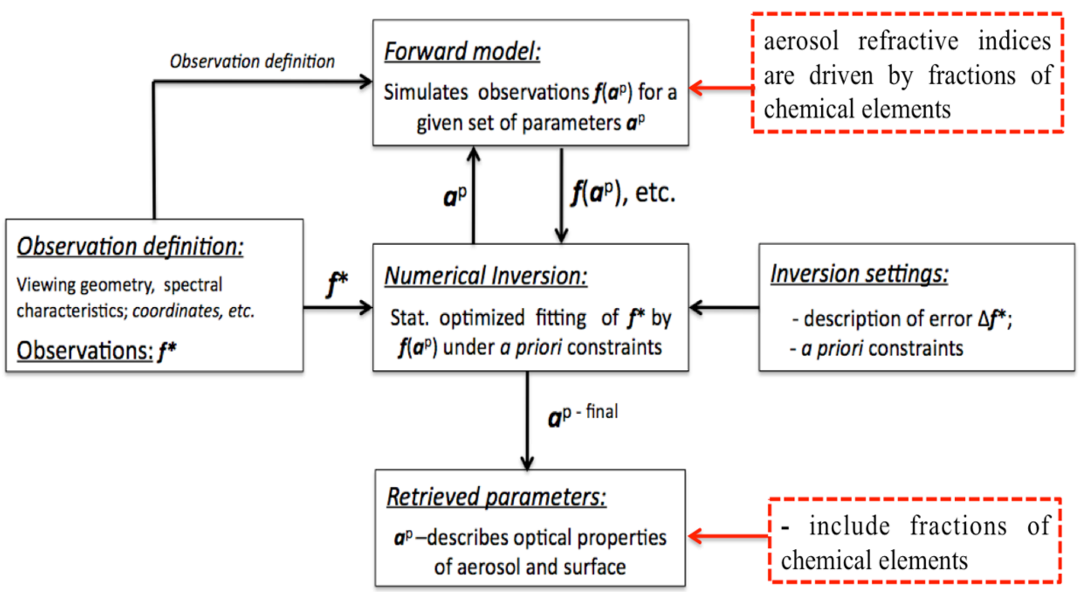
—without a prior knowledge of annual cycle.

GRASP retrieval of chemical compositions

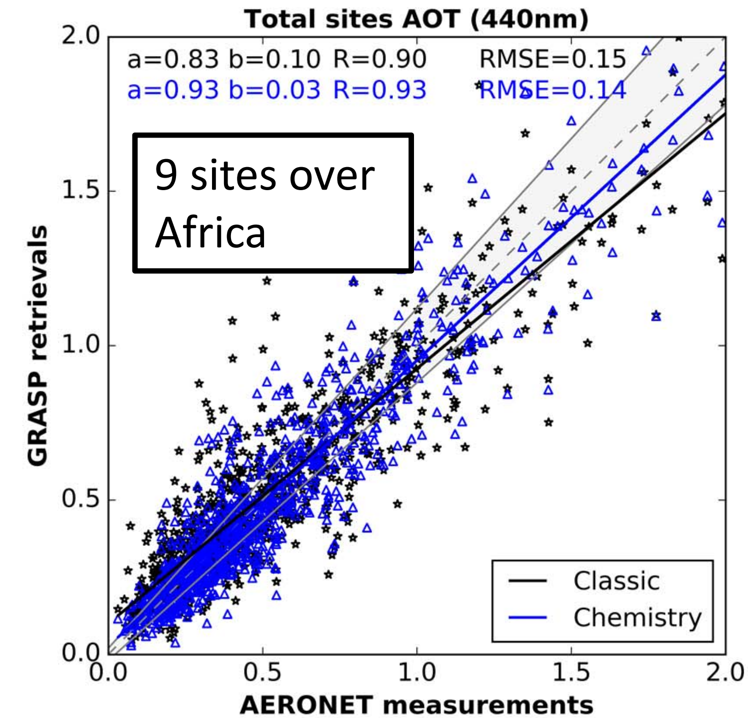
We add new elements and modify forward model in the GRASP algorithm

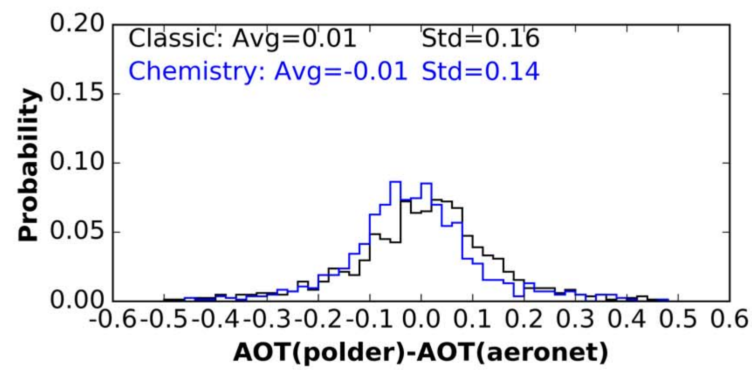
The comparisons of GRASP retrievals and AERONET measurements show that chemical approach is better than classic approach.

General structure of inversion algorithm



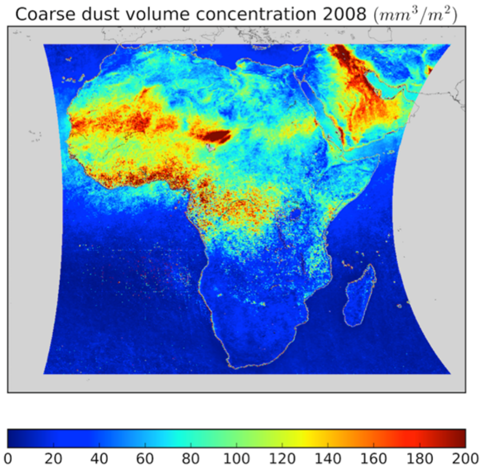
Contributed from Lei LI



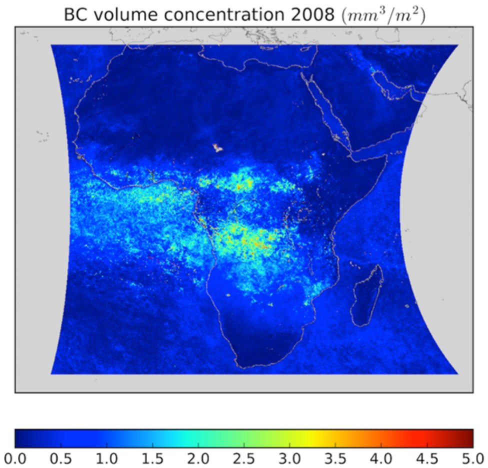


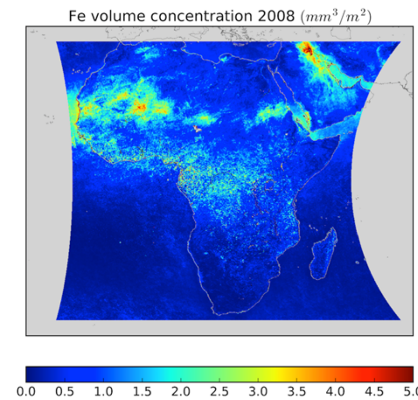
Volume concentration from PARASOL/GRASP-chemistry

Dust

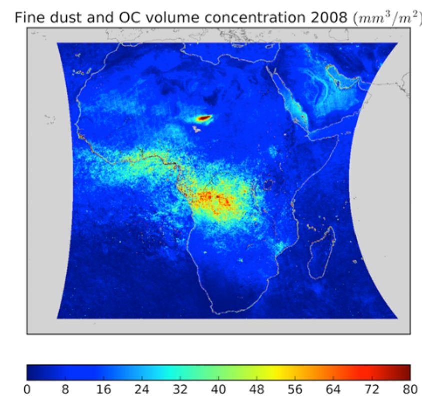


BC

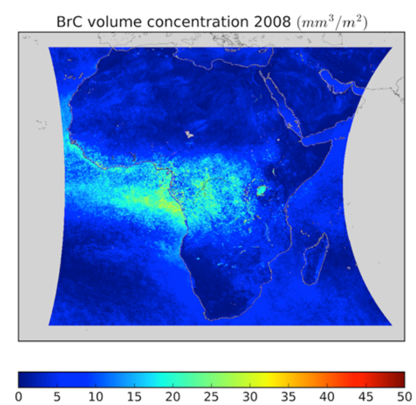




Fe

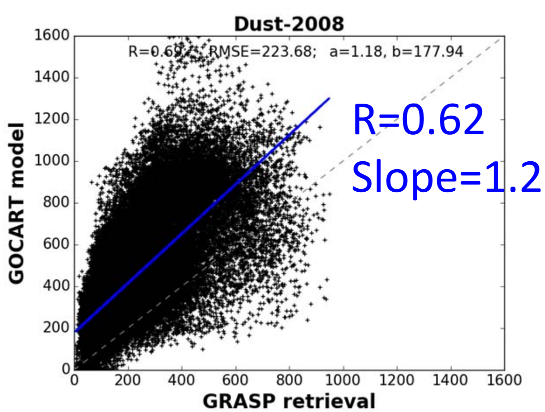


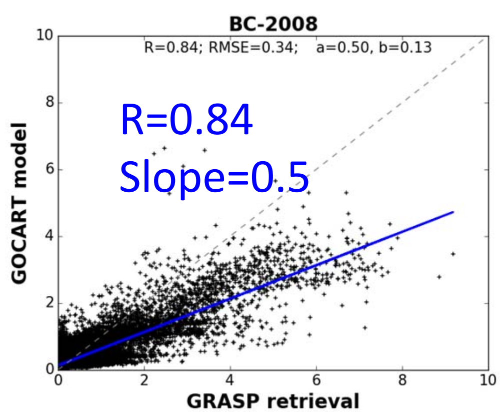
OC



BrC

GOCART *vs.* GRASP mass concentration





Mean over Africa

	Dust	BC
GOCART	291.7mg/m^2	0.58 mg/m^2
GRASP	184.6mg/m^2	0.90 mg/m^2

Perspectives: Retrieval emission from GRASP chemical products

- ✓ Retrieval of the aerosol emission from the GRASP new products of chemical components mass concentration (DU, BC, OC…).
  - ✓ Help to overcome the difficulties in estimate of the contribution from each components from spectral information of total AOD and AAOD

Conclusions and Perspectives

- The sensitivity test shows our method is capable to determine BC, OC and DU aerosol emission sources simultaneously from PARASOL/GRASP spectral AOD and AAOD with high accuracy.
- Highlights:
  - Emission retrieved from PARASOL/GRASP observations with weak constrain of a prior knowledge of emission distribution and strength.
  - Model posterior simulated aerosol properties (AOD, AAOD, SSA and AExp) with our emission database can fit well with independent measurements (AERONET, MODIS, OMI).
- Limitations:
  - We neglect the differences could be attributed to poorly modeled removal processes and model defined aerosol microphysical properties instead of emissions.
  - From optical view of satellite, our database can't distinguish between natural and anthropogenic sources.
- Future work:
  - Implement our satellite based aerosol emission database into other chemical transport models. Hope it can be helpful to improve aerosol simulation.
  - ……

Thanks for your attention !

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Acknowledge:

Labex CaPPA project
GRASP team
GEOS-Chem and adjoint GEOS-Chem team
AERONET team
GOCART team
…

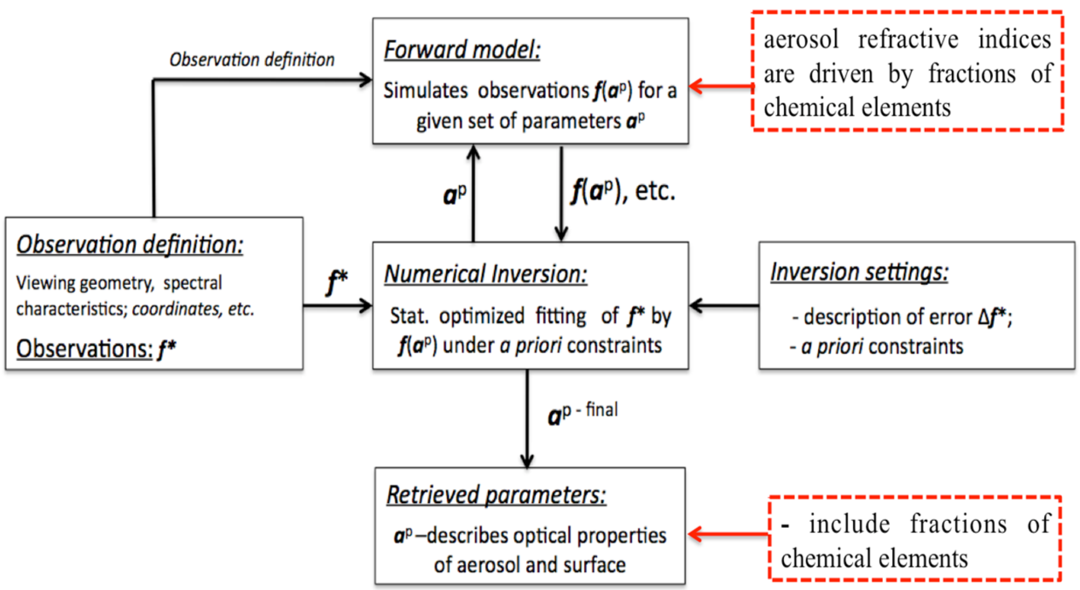
Physico-chemistry to refractive index conversion model

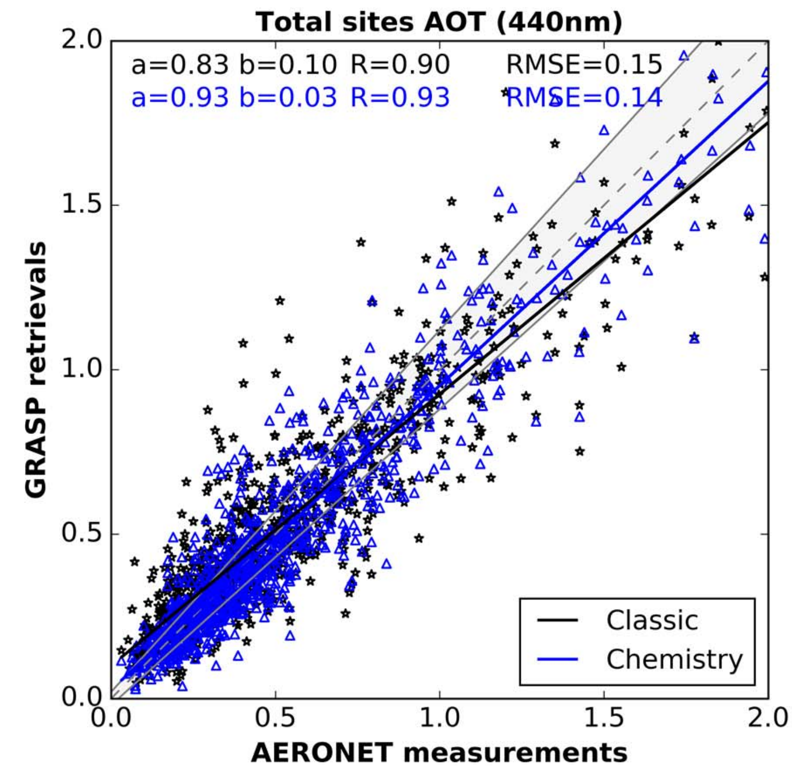
We add new elements and modify forward model in the GRASP algorithm

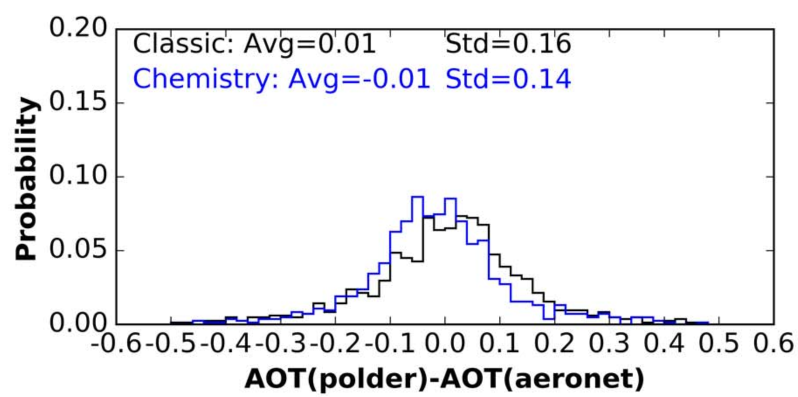
- ***size dependent*** *aerosol* composition
- **Fine mode: BC, BrC, Dust or WIOC (Water Insoluble Organic Carbon), Water like**
- **Coarse mode: Iron oxide(Fe), Dust, Water like**
- ***Volume-weighting mixture is assumed for mixture of the elements***

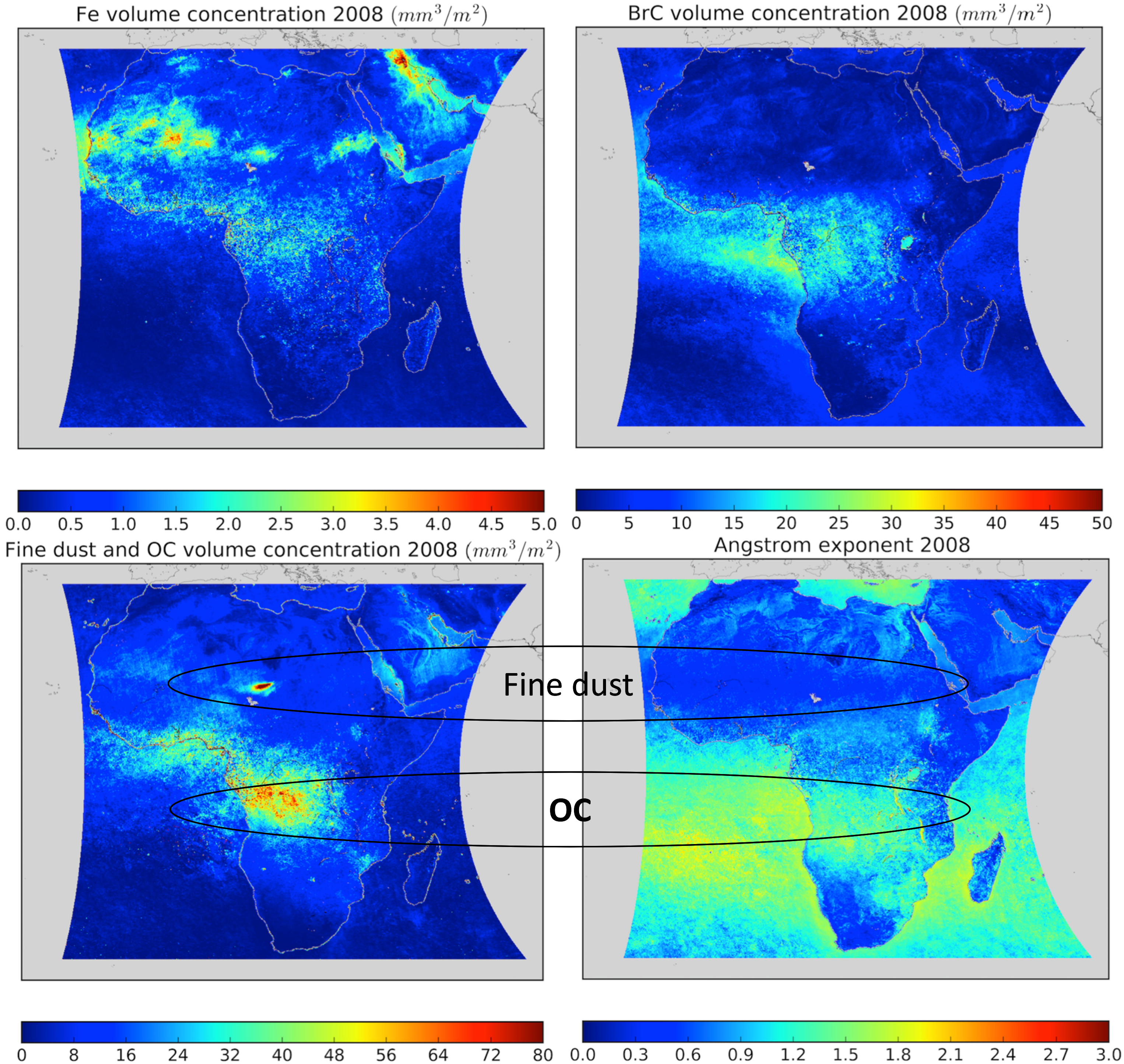
The comparisons of GRASP retrievals and AERONET measurements show that chemical approach is better than classic approach.

General structure of inversion algorithm









Fe volume concentration 2008 (mm^3/m^2)
0.0 0.5 1.0 1.5 2.0 2.5 3.0 3.5 4.0 4.5 5.0
BrC volume concentration 2008 (mm^3/m^2)
0 5 10 15 20 25 30 35 40 45 50
Fine dust and OC volume concentration 2008 (mm^3/m^2)
0 8 16 24 32 40 48 56 64 72 80
Angstrom exponent 2008
0.0 0.3 0.6 0.9 1.2 1.5 1.8 2.1 2.4 2.7 3.0
Fine dust
OC

*Table. Comparison of the annual mean DU, BC and OC emission flux between this study and other studies. The unit is Tg/yr. * The emission of this study is average from 2006 to 2011. + Here we simply account 10% BB aerosol for BC and 90% BB for OC.*

DU		BC		OC	
This study*	831	This study*	27.6	This study*	132.5
Prior GEOS-Chem	1345	Prior GEOS-Chem	11.0	Prior GEOS-Chem	47.2
IPCC (2013)	1000-4000	IPCC (2013)+	6.5-14.5	IPCC (2013) +	30.6-87.7
Dentener et al. (2006)	1678	Bond et al. (2004)	8.0	Bond et al. (2004)	33.0
Tanaka et al. (2006)	1877	Penner et al. (1993)	13.0-24.0		
Miller et al. (2004)	1019	Liousse et al. (1996)	12.0	Liousse et al. (1996)	73.0
Ginoux et al. (2004)	2073	Chin et al. (2009)	10.2	Chin et al. (2009)	61.8
Zender et al. (2003)	1490	Takemura et al. (2005)	15.98	Takemura et al. (2005)	105.7
Luo et al. (2003)	1654	Kim et al. (2008)	14.4	Kim et al. (2008)	54.4
Werner et al. (2002)	1060	Cooke and Wilson (1996)	14.0		
Huneeus et al. (2012)	1383	Huneeus et al. (2012)	15.0	Huneeus et al. (2012)	119.0

Emissions of BC, OA, and dust – BASE vs. CHEN_v2