We would like to thank the three referees for their time reviewing the manuscript, and for the helpful feedback provided. The detailed responses to all referees are provided below.

# **Reviewer #1:**

The paper describes a thorough evaluation of aerosol products retrieved by the GRASP algorithm (in different configurations) from POLDER-3/PARASOL. First, a comparison to AERONET for the full data set is presented. Second, a comparison with MODIS aerosol products is performed. It is concluded that the GRASP/Models AOD product is at least as good as (and probably better than) the MODIS AOD products and that the GRASP/HP product is superior for retrieving SSA and AE.

Overall, the paper is well written and the conclusions are sound. The part on the comparison with MODIS is quite detailed and sometimes a bit hard to follow (because of the comparison of 3 GRASP products with 3 MODIS products). I think this part can be shortened by removing the part of fine- and coarse mode AOD as I believe the AOD+AE comparison already tells the story.

I recommend publication of this paper after addressing my comments I added to the pdf file of the manuscript, most of which are minor.

# **Response:**

#### Dear Otto,

Thank you for the constructive and positive comments on our paper. Here are the point-by-point responses:

We agree that the Ångström exponent (AE) can qualitatively indicate the domination of fine or coarse model aerosol. At the same time, we would like to point out that quantitative analysis of fine/coarse mode contribution to total AOD is very challenging and unclear without fine mode AOD (AODF) and coarse mode AOD (AODC). Moreover, the AODF is a parameter of particular interest for various applied research targeting characterization of Air Quality and anthropogenic effects. For example, the Air Quality related studies of Wei et al. (2020) are focused on only AODF from POLDER and MODIS. In addition, the multi-angular polarimetry is known for high sensitivity to fine mode aerosol and the POLDER/Operational products provide AODF over land and ocean as the main "operational POLDER" products. Therefore, we consider beneficial to keep AODF and AODC analysis in our paper. We validated the POLDER/GRASP AODF with AERONET AODF, and intercompared with POLDER/Operational and MODIS over ocean. We believe that our analysis provide useful insights for the users of satellite AODF and AODC products.

Wei, Y., Z. Li, Y. Zhang, C. Chen, O. Dubovik, Y. Zhang, H. Xu, K. Li, J. Chen, H. Wang, B. Ge, C. Fan, "Validation of POLDER GRASP aerosol optical retrieval over China using SONET observations", J. Quant. Spectrosc. Radiait. Transfer, 246, 106931, 2020. doi:10.1016/j.jqsrt.2020.106931, 2020.

#### Main points:

Two comments I'd like to highlight here: - It seems that the GRASP/Models product has significantly less valid retrievals than the GRASP/HP product ( $\sim$ 31000 vs  $\sim$ 44000). What is the reason? Is the filter for GRASP/Models stricter? This is not clear from the text (in fact the opposite is suggested). May this be the reason for the better performance? Some discussion is needed here.

## **Response:**

Indeed, the different number of points and somewhat different approaches of quality filtering is one of the main shortcomings in our study. Reviewer #3 raised similar question. In fact, the post-processing flow (L1-L2-L3) was based on several attempts dictated by practical needs. These attempts provided us valuable inside but they could be fully evaluated only after full-scale validation. For example, the level 3 GRASP/HP and Optimized archives were generated and released much earlier than GRASP/Models. Also, we have done preprocessing of GRASP/Models over land at first and learned that screening was very conservative. Based on that we used less conservative screening for Models reprocessing over ocean. Once the products were released, they were used by many users, therefore, regenerating Level 3 products was not reasonable for this study. We are considering the harmonization of the all archives in future once time and resources allow that.

At the same time, we have looked at possible effect of applying tighter screening to HP and Optimized data. Our analysis showed that although stricter screening

somewhat improves the correlations, it doesn't change conceptually the results of validations. For example, it does not improve the BIAS which is considered as a main issue for these data sets. Some explanations of this aspect were added in the Sect 2.4 and Sect 3.1 as follow.

"For GRASP/Models product we did not use any filter, because a stricter quality assurance filter has been applied in GRASP/Models products generation from L1 to L2 and L3 than for other GRASP datasets. In principle, the post-processing of all PARASOL/GRASP products was done in similar ways. At the same time, the L3 products were prepared and released not at the same time. For example, the L3 GRASP/HP and Optimized archives were generated and released much earlier than GRASP/Models. Therefore, the post-processing and quality screening approaches used for different data archives are not exactly the same. Unfortunately, most of the differences were identified after the release of the products, its extensive use and the full-scale validation. In these regards, the harmonization of the all archives is likely to be done in future, but it will lead in release of new products."

"As described in section 2.4, the different post-processing scheme resulted in the difference for matched points between GRASP/Optimized, GRASP/HP and GRASP/Models. It can be noticed that applying a much stricter filter may improve the overall correlation against AERONET AOD for GRASP/HP and GRASP/Optimized products, but leads to significant loss of points and, most importantly, do not improve the BIAS, which is considered as a main issue for them."

The evaluation puts large focus on the correlation coefficient when comparing the performance of different products. This is not always a good metric because it is heavily influenced by the range, i.e. a limited number of points at the end of the range can have large effect on the correlation. I recommend to put more emphasis on other metric such as RMSE and MAE (Mean Absolute Error).

# **Response:**

The idea of our analysis was to provide comprehensive characterization of the observed relationships, rather than to focus on one selected parameter. Therefore, we provided many different parameters describing the relationship and did not intend to focus on correlation coefficient. Thanks for the suggestion about evaluation metrics!

We have revised the text to make more comprehensive discussion of other correlation parameters including RMSE and BIAS.

At the same time, we would like to note that RMSE and BIAS also have limitations in characterizing quality of agreement. For example, RMSE is always much lower for the dataset that include mainly small values of AOD. Therefore, the retrievals failing to report the retrieval at larger AODs have smaller RMSE than those that provide AODs with larger values.

## Minor points and grammar:

Line 103: Please also cite Mishchenko and Travis, JGR, 1997 doi:10.1029/96JD02425 and Hasekamp and Landgraf, Appl. Opt., 2007, https://doi.org/10.1364/AO.46.003332)

# **Response:**

Corrected.

Line 139: 910 nm Response: Corrected.

Line 159: Mention that also other MAP algorithms have been developed / are being devloped: SRON (Hasekamp et al., J. Geophys. Res., 116, D14204, https://doi.org/10.1029/2010JD015469, 2011: Fu and Hasekamp https://doi.org/10.5194/amt-11-6627-2018, 2018) JPL (Xu et al.. https://doi.org/10.1002/2017jd026776, 2017; https://doi.org/10.3390/rs11070746, 2019), LaRC (Stamnes et al., https://doi.org/10.1364/AO.57.002394, 2018.) All these algorithms follow a similar principle of online RT calculations and no restriction to aerosol models.

## **Response:**

We fully agree that the text of article didn't mention other advanced algorithms and therefore could be misleading. We added corresponding discussion in Sect. 2.1 as below.

"A unique aspect of GRASP is that it can perform radiative transfer (RT) computations fully accounting for multiple interactions of the scattered solar light in

the atmosphere online without the use of traditional LUTs. Several other algorithms of new generation have been or being developed for interpretation of MAP observation use the online RT calculations and implement retrieval as a search in continuous space of solution e.g. Hasekamp et al., 2011, Xu et al., 2017, 2019, Fu and Hasekamp, 2018, Gao et al., 2018, Stamnes et al., 2018, Di Noia et al., 2019. Nonetheless, at present GRASP was the only algorithm that has been used to generate aerosol products for full archive of POLDER observations (Dubovik et al., 2019)."

- Dubovik, O., Li, Z., Mishchenko, M. I., Tanré, D., Karol, Y., Bojkov, B., Cairns, B., Diner, D. J., Espinosa, W. R., Goloub, P., Gu, X., Hasekamp, O., Hong, J., Hou, W., Knobelspiesse, K. D., Landgraf, J., Li, L., Litvinov, P., Liu, Y., Lopatin, A., Marbach, T., Maring, H., Martins, V., Meijer, Y., Milinevsky, G., Mukai, S., Parol, F., Qiao, Y., Remer, L., Rietjens, J., Sano, I., Stammes, P., Stamnes, S., Sun, X., Tabary, P., Travis, L. D., Waquet, F., Xu, F., Yan, C. and Yin, D.: Polarimetric remote sensing of atmospheric aerosols: Instruments, methodologies, results, and perspectives, J. Quant. Spectrosc. Radiat. Transf., 224, 474–511, doi:10.1016/J.JQSRT.2018.11.024, 2019.
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- Hasekamp, O. P., Litvinov, P. and Butz, A.: Aerosol properties over the ocean from PARASOL multiangle photopolarimetric measurements, J. Geophys. Res., 116(D14), D14204, doi:10.1029/2010JD015469, 2011.
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- Stamnes, S., Hostetler, C., Ferrare, R., Burton, S., Liu, X., Hair, J., Hu, Y.,

Wasilewski, A., Martin, W., van Diedenhoven, B., Chowdhary, J., Cetinić, I., Berg, L. K., Stamnes, K. and Cairns, B.: Simultaneous polarimeter retrievals of microphysical aerosol and ocean color parameters from the "MAPP" algorithm with comparison to high-spectral-resolution lidar aerosol and ocean products, Appl. Opt., 57(10), 2394, doi:10.1364/ao.57.002394, 2018.

- Xu, F., van Harten, G., Diner, D. J., Kalashnikova, O. V., Seidel, F. C., Bruegge, C. J. and Dubovik, O.: Coupled retrieval of aerosol properties and land surface reflection using the Airborne Multiangle SpectroPolarimetric Imager, J. Geophys. Res., 122(13), 7004–7026, doi:10.1002/2017JD026776, 2017.
- Xu, F., Diner, D. J., Dubovik, O. and Schechner, Y.: A correlated multi-pixel inversion approach for aerosol remote sensing, Remote Sens., 11(7), doi:10.3390/rs11070746, 2019.

Line 188: SPEXone

# **Response:**

Corrected.

Line 191: It is important to mention in the introduction that advanced MAP aerosol products from POLDER-3 (from GRASP and SRON algorithms ) have already been used for emission estimates (Chen et al, https://doi.org/10.5194/acp-18-12551-2018, 2018; https://doi.org/10.5194/acp-19-14585-2019, 2019); data assimilation (Tsikerdekis et al., https://doi.org/10.5194/acp-2020-468), estimating the direct radtive effect of aerosols (Lacagnina et al., https://doi.org/10.1002/2015jd023501, 2015. https://doi.org/10.1002/2016jd025706, 2017) and radiative forcing due to aerosol-cloud interactions (Hasekamp et al., https://doi.org/10.1038/s41467-019-13372-2, 2019).

This shows the potential of such advanced MAP products.

## **Response:**

We now provide discussion as follow.

"Several studies have shown the potential of advanced MAP aerosol products, for example, PARASOL/GRASP results have been adopted to estimate global aerosol emissions (Chen et al., 2018; 2019), PARASOL/SRON products have been used for data assimilation (Tsikerdekis et al., 2020), estimation of aerosol direct radiative effect (Lacagnina et al., 2015; 2017), and the radiative forcing due to aerosol-cloud interactions (Hasekamp et al., 2019b)."

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  F., Fuertes, D., Litvinov, P., Li, L., Lopatin, A., Hu, Q. and Torres, B.:
  Constraining global aerosol emissions using POLDER/PARASOL satellite
  remote sensing observations, Atmos. Chem. Phys., 19(23), 14585–14606,
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- Chen, C., Dubovik, O., Henze, D. K., Chin, M., Lapyonok, T., Schuster, G. L., Ducos, F., Fuertes, D., Litvinov, P., Li, L., Lopatin, A., Hu, Q. and Torres, B.: Constraining global aerosol emissions using POLDER/PARASOL satellite remote sensing observations, Atmos. Chem. Phys., 19(23), 14585–14606, doi:10.5194/acp-19-14585-2019, 2019.
- Hasekamp, O. P., Gryspeerdt, E. and Quaas, J.: Analysis of polarimetric satellite measurements suggests stronger cooling due to aerosol-cloud interactions, Nat. Commun., 10(1), 1–7, doi:10.1038/s41467-019-13372-2, 2019b.
- Lacagnina, C., Hasekamp, O. P., Bian, H., Curci, G., Myhre, G., van Noije, T., Schulz, M., Skeie, R. B., Takemura, T. and Zhang, K.: Aerosol single-scattering albedo over the global oceans: Comparing PARASOL retrievals with AERONET, OMI, and AeroCom models estimates, J. Geophys. Res., 120(18), 9814–9836, doi:10.1002/2015JD023501, 2015.
- Lacagnina, C., Hasekamp, O. P. and Torres, O.: Direct radiative effect of aerosols based on PARASOL and OMI satellite observations, J. Geophys. Res., 122(4), 2366–2388, doi:10.1002/2016JD025706, 2017.
- Tsikerdekis, A., Schutgens, N. A. J. and Hasekamp, O. P.: Assimilating aerosol optical properties related to size and absorption from POLDER/PARASOL with an ensemble data assimilation system, Atmos. Chem. Phys. Discuss., in review, doi:10.5194/acp-2020-468, 2020.

Line 210: There are also other algorithms that do that (see my comment above). **Response:** 

#### Corrected.

Line 307: In Introduction together with other MAP aerosol applications (see above). **Response:** 

#### Corrected.

Line 416: I would say for SSA/AAOD it is more comparison than validation because also AERONET has large uncertainties for these properties.

#### **Response:**

We agree that SSA and AAOD provided by AERONET have substantial uncertainties in some situations (such as the low-AOD case). At the same time, we consider that AERONET, so far, provides overall the most comprehensive and reliable SSA and AAOD data among all available data sources. Since AERONET retrievals rely on direct Sun observation for getting AOD, they have serious advantages over satellite data for constraining aerosol absorption retrieval.

At the same time, we do not consider our POLDER paper as a right place for the discussions, we rewrote the sentence as follows to avoid the discussion: "AAOD and SSA products are chosen as references for satellite products comparison and evaluation."

# Line 430: How to go from 6km to 0.1 degree?

## **Response:**

The gdalwarp regridding technique (https://gdal.org/programs/gdalwarp.html) is used to generate 0.1 degree products from original ~6 km retrieval. The algebraic mean is then calculated from all pixels in a grid box.

#### Line 440: How to interpret these numbers (residual)?

#### **Response:**

The relative residual is the root mean square of relative error in fitting the measurements by the algorithm.

Line 441: Not clear. Actually the GRASP/Models results seem most heavily filtered as there are much less points in the validation plots.

## **Response:**

Rather elaborated filtering scheme was used to generate GRASP/Models L3 products. Hence, no additional filtering was applied to L3 GRASP/Models 0.1degree products in the validation considerations. Line 450: So, this is 9 0.1 degree pixels?

# **Response:**

Not exactly. For POLDER/GRASP and MODIS DT, DB and MAIAC\_0.1, it is 9 0.1 degree pixels. While for MAIAC\_0.01, 9 0.01 degree pixels are used, and 9 18.5 km pixels are for POLDER/Operational products. We now clarify this in the text.

#### Line 461: Why? Indications for clouds?

# **Response:**

The purpose of this criterion is to remove some evident outliers, which stand out within 3x3 or 9x9 windows. It is proven to be helpful for validation. It is quite possible that the high variability in 3x3 or 9x9 windows can be related to clouds, but further investigations for supporting this idea are needed.

Line 480: I would say the AERONET uncertainty should be added quadratically ....

## **Response:**

We agree that if we consider the statistical rule of adding standard deviations the quadratic addition should be done. However, GCOS criteria seem to be a criterion defined based on practical considerations rather than rigorous statistical consideration. In this regards, we simply followed the common practice and have adopted the GCOS requirements,  $GCOS = max(\pm 0.04, \pm 0.1AOD)$ , following the latest Aerosol\_cci study (Popp et al., 2016).

#### Line 489: Why just 2008?

# **Response:**

The year 2008 was chosen as an example year to comprehensive evaluation of the consistencies and differences between POLDER/GRASP and MODIS aerosol products. The observations during 2008 year contain generally good observational statistics and all types of aerosol are clearly present. The year has also been used as a reference in many evaluation studies, e.g., in Aerosol\_cci study.

Line 505: But there are much less points. **Response:** 

Yes, this is correct, GRASP/Models product contains less point than GRASP/Optimized and GRASP/HP due to different filter scheme used for generation of L3 0.1degree products. This aspect has already been discussed above and a discussion is added in the Sect. 2.4 and Sect 3.1 of the revised manuscript.

Line 543: Do fine and coarse mode have the same definition on GRASP and AERONET?

## **Response:**

The separation of the fine and coarse modes in AERONET and GRASP are not exactly the same, but they provide very close results in case then both modes are separated by distinct minimum.

Line 582: So, is GRASP/Models really better? Or is it a 'lucky' compensation of errors?

## **Response:**

In terms of spectral AOD products, GRASP/Models show better agreement with AERONET measurements in many senses. The biggest problem of GRASP/Optimized and GRASP/HP AOD products is a distinct BIAS. At the same time, the validation of AODF and AODC indicates that the BIAS for GRASP/Optimized and GRASP/HP comes mainly from the coarse mode, and the AODF provided by GRASP/Optimized and GRASP/HP has almost no BIAS and show statistic that even better than AODF from GRASP/Models. This observation suggests that the approach of GRASP/Optimized and GRASP/HP can likely be improved in the future.

By the way, this result shows also the importance of keeping the section of fine and coarse mode validation for helping to improve understanding of the overall retrieval performance.

Line 592: But you compare against AERONET L2 which only includes AOD > 0.4. So, I don't understand the low AOD filter over ocean. because by comparing to AERONET L2 only large AOD cases will be included in the end. Or am I missing something?

#### **Response:**

Yes, we are comparing against AERONET L2 inversion data, which includes only AERONET AOD>0.4. Here we use additional filter for satellite AOD (Land: AOD 443 nm>0.3; Ocean: AOD 443 nm>0.02).

#### Line 655: What do you mean by this?

#### **Response:**

We rewrote the sentence as follows. "MAIAC products cover some land-containing ocean tiles, however due to limited coverage of these retrievals we do not consider MAIAC ocean products here."

Line 717: It would be rather straightforward to compure GRASP AOD at 470 and 660 so that the exact same quantity can be compared.

#### **Response:**

In the paper, we intended to present the standard AE products validation for each dataset. Hence, we decided to use different wavelengths to compute AE for POLDER/GRASP and MODIS. Figure R1 shows the comparison of POLDER/GRASP AE (470/660) against with AERONET AE (470/660). This comparison is done by interpolating both POLDER and AERONET AOD to 470 and 660 nm based on the nearest available wavelengths. It presents similar performance with GRASP AE (440/870) in Figure 11.



Figure R1. Validation of POLDER/GRASP (a. GRASP/Optimized, b. GRASP/HP and c. GRASP/Models) AE over land in 2008.

Line 899: decrease?

### **Response:**

Thanks. It should be 'decrease'. It was revised.

Line 948: I would say the other statistical parameters are more important to discuss than the correlation.

#### **Response:**

In the revised manuscript, we have adjusted the discussion with including more comprehensive discussion of other parameters, e.g. R, RMSE, BIAS, GCOS, etc.

Line 1055: This needs more discussion. The bias between GRASP/Models and GRASP/HP is larger for most of the globe than between GRASP/HP and DT, right? This is quite unexpected given the AERONET comparison.

#### **Response:**

In Figure 24, indeed the differences between GRASP/Models and GRASP/HP are larger than DT and GRASP/HP. Table 18 confirms the difference (GRASP/Models - GRASP/HP) is 0.12 over land, and DT - GRASP/HP is 0.11; over ocean, the difference (GRASP/Models - GRASP/HP) is 0.35, and DT - GRASP/HP is 0.25. This is due to the phenomenon that GRASP/Models tend to overestimate AE for coarse particles (AE $\sim$ 1.0) (see Figure 3). While in terms of correlation, GRASP/Models and GRASP/HP (Table 18) agree well (R>0.7) over both land and ocean, which can be interpreted for agreement in qualitatively indicating fine or coarse model aerosol. Overall, GRASP/Models can be improved by adjusting adopted aerosol models specifically for coarse mode models.

Line 1124: It is important to highlight the better AOD performance of GRASP/Models than the other GRASP versions before going to the comparison with other data sets. Namely, the better performance holds for GRASP/Models but not always for the other versions.

#### **Response:**

We now clarify it in the text as follow.

"the PARASOL spectral products including AOD for six wavelengths in the range 443 to 1020 nm agree well with AERONET AOD measurements, e.g. for PARASOL/Models AOD correlation coefficients R are  $\geq 0.86$  over land and  $\geq 0.94$  over ocean with BIAS not exceeding 0.01 over land and 0.02 over ocean for all wavelengths. PARSOL/Optimized and PARASOL/HP also show good agreement with AERONET for spectral AOD, however they have non-negligible bias ~0.05-0.07 spectrally."

# **Reviewer #2:**

#### Dear Kirk,

We would like to thank you for the constructive and positive comments on our paper. Please find our responses below.

First and foremost, I'm not sure this manuscript is within the scope of ESSD. The "Aims and scope" portion of the ESSD website (https://www.earth-systemsciencedata.net/about/aims and scope.html) says: "Earth System Science Data (ESSD) is an international, interdisciplinary journal for the publication of articles on original research data (sets), furthering the reuse of high quality data of benefit to Earth system sciences. The editors encourage submissions on original data or data collections which are of sufficient quality and have the potential to contribute to these The journal maintains sections for regular length articles, brief aims. communications (e.g. on additions to data sets) and commentaries, as well as review articles and special issues. Articles in the data section may pertain to the planning, instrumentation, and execution of experiments or collection of data. Any interpretation of data is outside the scope of regular articles. Articles on methods describe nontrivial statistical and other methods employed (e.g. to filter, normalize, or convert raw data to primary published data) as well as nontrivial instrumentation or operational methods. Any comparison to other methods is beyond the scope of regular articles." I would think that an ESSD style manuscript on this topic would

simply describe the various GRASP algorithms (briefly) and where they are archived. The majority of the paper is indeed comparisons to other data sets. I think it is far more appropriate for Atmospheric Measurement Techniques, for example (to pick another Copernicus Journal). I imagine this is something the editor needs to weigh in upon, but I wouldn't look to ESSD for this type of manuscript.

#### **Response:**

Multi-angular polarimetry (MAP) is always considered ideal for comprehensive retrieval of aerosol properties. GRASP algorithm was developed originally for operational processing of MAP measurements (Dubovik et al., 2011; 2014). The goal of this study is to announce the release of three archives of multi-angular polarimetry POLDER aerosol products processed by GRASP algorithm and provide comprehensive evaluation of these products against ground-based AERONET dataset, and popular MODIS aerosol products from DT, DB and MAIAC algorithms. For example, we found out that the quality of AOD retrieval from MAP (e.g. POLDER) is at least comparable to those of MODIS like imagers. In addition, we show that the MAP observations provide more information on detailed aerosol properties, e.g. spectral fine/coarse AOD, AE, as well as aerosol absorption properties such as AAOD and SSA. In this way, we assessed the potential of MAP sensors for aerosol monitoring. These both aspects are not surprising and were already discussed intensively in aerosol community. At the same, the absence of actual product from MAP sensors has often used as an argument for suggesting some overstatement of MAP potential. In these regards, our paper is aimed to answer this pessimism.

Several of our colleagues and co-authors suggested publishing our paper in new ESSD journal. Additionally, we were also inspired by your paper (Knobelspiesse et al., 2020) published in Earth System Science Data descripting the ACEPOL (Aerosol Characterization from Polarimeters and Lidar) field campaign, both of them show advances for aerosol characterization by utilizing the new era of MAP measurements from different perspectives. After additional consideration, we admit publishing our paper in other journals could be appropriate, but we remain convinced that this manuscript is rather appropriate for the Earth System Science Data. In addition, given the fact, that it was already exposed in open discussion and received several reviews, we prefer to continue with this journal. Secondly, about the length. The manuscript review version is 108 pages long with 23 tables and 28 figures. You've chosen an extensive set of data to compare and contrast the various versions of POLDER data to the various versions of MODIS and then again to AERONET. What I'm missing is a concise set of objectives and how those are met. While I'm sure they are in the manuscript, they are lost among the noise. Because of the scale of the task, you need to be creative in finding ways to condense all of this analysis into something that easily and simply supports your work. Given the above two points, you may consider splitting this manuscript. For example, you could make an ESSD manuscript that describes basics of the dataset and its creation and archive location. It would point to one (or more) manuscripts that contain analysis. One could be on continuity with MODIS (and VIIRS?) and another on the full set of PARASOL products and comparison to AERONET. This is just a suggestion.

#### **Response:**

We agree that the manuscript is a long and that for the reader may be not easy to follow all details of the manuscript. In order to address that, we have revised the manuscript by combining all comments from reviewers, and trying to make it more readable. Generally, there are 3 main parts of this manuscript, (1) validation three archives (Optimized, HP and Models) PARASOL/GRASP products (spectral AOD, AE, AODF, AODC, AAOD and SSA) against AERONET data for entire PARASOL 2005-2013; (2) comparison of results obtained from validation of PARASOL (GRASP and Operational) and MODIS (DT, DB and MAIAC) aerosol products against AERONET in year 2008; (3) Inter-comparison satellite products at global pixel-to-pixel scale. The first part was on the full set of PARASOL products and comparison to AERONT. The second part tried to compare PARASOL and MODIS aerosol products by validating with AERONET follow the same criteria. The third part was inter-comparing PARASOL and MODIS aerosol products over globe at pixel level. We agree that they can be split in different papers but the separation of the materials of the paper is very difficult, because these three parts are quite complimentary. We feel that having the 3 parts together make the story more complete and after considerations we prefer to not split the paper.

Since the core of this manuscript is comparisons of other datasets, the choice of statistical metrics is very important. Table 3, for example, shows the use of nine

different metrics, although Pearson's linear correlation coefficient is most commonly employed in the text. Unfortunately, some of these metrics, and especially the linear correlation coefficient, are not suitable for use on non-gaussian distributed data. The correlation coefficient is an expression of association, not agreement (Altman and Bland, 1983 and Bland and Altman, 1986), and is subject to numerical distribution, outliers, and sample range. So, for example, the R values for SSA are lower, but is that because of the lower success of the PARASOL retrieval (what you want to know) or because of the truncated numerical distribution of SSA which makes it non gaussian distributed? Additionally, what threshold of R can be considered a success? To that end, I think your metric for percentage within the GCOS requirements is a much more appropriate measure, and should instead be emphasized, although you need to take care to account for measurement uncertainty in both POLDER and MODIS or AERONET. Seegers et al. 2018 is a nice overview of these issues in ocean color data products, but equally appropriate here. They also identify regression slope and root mean square error as problematic, while noting that mean bias (which you use) and mean absolute error as appropriate. Bland and Altman suggest something similar, also recommending the pairwise mean bias and the "limits of agreement" which is similar to the mean absolute error. Variable measurement uncertainty can also be incorporated into these techniques (Knobelspiesse et al, 2019) which addresses the salient question "Do measurements agree to within stated uncertainties?" Ultimately, you should revise (and perhaps simplify) the metrics you use to assess your results.

Altman, D. G. and Bland, J. M.: Measurement in medicine: the analysis of method comparison studies, The statistician, 307–317, 1983.

Bland, J. M. and Altman, D.: Statistical methods for assessing agreement between two methods of clinical measurement, The lancet, 327(8476), 307–310, 1986.

Knobelspiesse, K., Tan, Q., Bruegge, C., Cairns, B., Chowdhary, J., van Diedenhoven, B., Diner, D., Ferrare, R., van Harten, G., Jovanovic, V., Ottaviani, M., Redemann, J., Seidel, F., and Sinclair, K.: Intercomparison of airborne multi-angle polarimeter observations from the Polarimeter Definition Experiment, Appl. Optics, 58(3), 650–669, 2019.

Seegers, B. N., Stumpf, R. P., Schaeffer, B. A., Loftin, K. A., and Werdell, P. J.: Performance metrics for the assessment of satellite data products: an ocean color case study, Optics express, 26(6), 7404–7422, 2018.

#### **Response:**

We fully agree that to choosing the adequate statistic metrics is very challenging. Therefore, for addressing this challenge we have presented many parameters at the same. In our understanding this approach allows us to have more comprehensive evaluation of the comparison results. Indeed, each single criterion has some limitations. For example, we agree that GCOS requirement is a good measure for AOD comparison, however, the total GCOS value tends to bias to small AOD, since >70% cases are coming from AOD<0.2. In Figures 7 and 8, GRASP/Models, MODIS DT, DB and MAIAC are all showing GCOS>45% for all AOD cases; while for AOD>0.2, most of them having GCOS<30%. The root mean square error (RMSE) also tends to be smaller for the products dominated by the results at lower AOD. The BIAS may be misleading in cases when many deviations with opposite sign are added together. For example, it is often a case for AE. In Figure 3, GRASP/Models tend to overestimate of small AE and underestimate high AE, and total BIAS is close to ~zero, which is smaller than GRASP/Optimized and GRASP/HP. Another example, it the Table 10, where for very bright surfaces, many retrievals have positive bias that is compensated by the negative bias at higher AODs. Thus, in the revised version of the paper, we have tired not to focus the discussion on a single parameter throughout the manuscript. Also, we wanted to make sure to provide all parameters, such as correlation coefficients, that are traditionally used in satellite comparisons. This helps us compare our results with published ones. In addition, following your recommendations, we have revised the expression to emphasize on all evaluation metrics, e.g. R, RMSE, BIAS, Slope, Offset and GCOS.

Co-author Sayer has a publication about the numerical distribution of AOD and its impact on averages of data which is relevant to your matchup methodology and your use of Level 3 products. Were you calculating arithmetic or geometric means? I assume they are arithmetic since you don't mention otherwise, however this can in some cases cause an artificial bias in the results.

Sayer, A. M. and Knobelspiesse, K. D.: How should we aggregate data? Methods accounting for the numerical distributions, with an assessment of aerosol optical depth, Atmos. Chem. Phys., 19(23), 15023–15048, https://doi.org/10.5194/acp-19-15023-2019, 2019.

## **Response:**

Yes, the L3 0.1 degree products were calculating using arithmetic mean and gdalwarp regridding technique (https://gdal.org/programs/gdalwarp.html). We have included this information in the text. As the spatial resolution is fine (compared to typical satellite composites at 1 degree), the arithmetic vs. geometric differences (as discussed in Sayer & Knobelspiesse, 2019) is likely significantly smaller for the present case.

I'm a little surprised that you make no mention of the retrieval algorithms for PARASOL (Hasekamp et al. 2011). In the beginning of section 2.1 you mention "A unique aspect of GRASP is that it can perform radiative transfer (RT) computations fully accounting for multiple interactions of the scattered solar light in the atmosphere, and that it can perform it online without the use of traditional LUTs." You show that GRASP is an extremely power retrieval algorithm, but it is certainly not unique in its use of iterative RT computations, and I find it problematic that you make this claim and do not mention similar algorithms. Without acknowledging that there are other algorithms, even for POLDER, the manuscript sounds more like a sales pitch for GRASP and less an dispassionate piece of peer reviewed literature. In addition to Hasekamp et al, 2011, which is applied to POLDER/PARASOL, many others come to mind including those listed below.

Di Noia, A., Hasekamp, O. P., Wu, L., van Diedenhoven, B., Cairns, B., and Yorks, J. E.: Combined neural network/Phillips–Tikhonov approach to aerosol retrievals over land from the NASA Research Scanning Polarimeter, Atmos. Meas. Tech., 10(11), 4235–4252, https://doi.org/10.5194/amt-10-4235-2017, 2017.

Fu, G. and Hasekamp, O.: Retrieval of aerosol microphysical and optical properties over land using a multimode approach, Atmos. Meas. Tech., 11(12), 6627–6650 , https://doi.org/10.5194/amt-11-6627-2018, 2018.

Gao, M., Zhai, P.-W., Franz, B., Hu, Y., Knobelspiesse, K., Werdell, P. J., Ibrahim, A., Xu, F., and Cairns, B.: Retrieval of aerosol properties and water-leaving reflectance from multi-angular polarimetric measurements over coastal waters, Optics express, 26(7), 8968–8989, 2018.

Hasekamp, O. P. and Landgraf, J.: Retrieval of aerosol properties over land surfaces: capabilities of multiple-viewing-angle intensity and polarization measurements, Appl. Optics, 46(16), 3332–3344 , 2007.

Hasekamp, O. P., Litvinov, P., and Butz, A.: Aerosol properties over the ocean from PARASOL multiangle photopolarimetric measurements, J. Geophys. Res, 116(D14), D14204, 2011.

# **Response:**

We fully agree with this criticism and revised the text as below.

"A unique aspect of GRASP is that it can perform radiative transfer (RT) computations fully accounting for multiple interactions of the scattered solar light in the atmosphere online without the use of traditional LUTs. Several other algorithms of new generation have been or being developed for interpretation of MAP observation use the online RT calculations and implement retrieval as a search in continuous space of solution e.g. Hasekamp et al., 2011, Xu et al., 2017, 2019, Fu and Hasekamp, 2018, Gao et al., 2018, Stamnes et al., 2018, Di Noia et al., 2019. Nonetheless, at present GRASP was the only algorithm that has been used to generate aerosol products for full archive of POLDER observations (Dubovik et al., 2019)."

- Dubovik, O., Li, Z., Mishchenko, M. I., Tanré, D., Karol, Y., Bojkov, B., Cairns, B., Diner, D. J., Espinosa, W. R., Goloub, P., Gu, X., Hasekamp, O., Hong, J., Hou, W., Knobelspiesse, K. D., Landgraf, J., Li, L., Litvinov, P., Liu, Y., Lopatin, A., Marbach, T., Maring, H., Martins, V., Meijer, Y., Milinevsky, G., Mukai, S., Parol, F., Qiao, Y., Remer, L., Rietjens, J., Sano, I., Stammes, P., Stamnes, S., Sun, X., Tabary, P., Travis, L. D., Waquet, F., Xu, F., Yan, C. and Yin, D.: Polarimetric remote sensing of atmospheric aerosols: Instruments, methodologies, results, and perspectives, J. Quant. Spectrosc. Radiat. Transf., 224, 474–511, doi:10.1016/J.JQSRT.2018.11.024, 2019.
- Fu, G. and Hasekamp, O.: Retrieval of aerosol microphysical and optical properties over land using a multimode approach, Atmos. Meas. Tech., 11(12), 6627–6650, doi:10.5194/amt-11-6627-2018, 2018.
- Gao, M., Zhai, P.-W., Franz, B., Hu, Y., Knobelspiesse, K., Werdell, P. J., Ibrahim, A., Xu, F. and Cairns, B.: Retrieval of aerosol properties and water-leaving reflectance from multi-angular polarimetric measurements over coastal waters, Opt. Express, 26(7), 2973–2984, doi:doi.org/10.1364/OE.26.008968, 2018.
- Hasekamp, O. P. and Landgraf, J.: Retrieval of aerosol properties over land surfaces: Capabilities of multiple-viewing-angle intensity and polarization measurements, Appl. Opt., 46(16), 3332–3343, doi:10.1364/AO.46.003332, 2007.

Hasekamp, O. P., Litvinov, P. and Butz, A.: Aerosol properties over the ocean from

PARASOL multiangle photopolarimetric measurements, J. Geophys. Res., 116(D14), D14204, doi:10.1029/2010JD015469, 2011.

- Di Noia, A., Hasekamp, O. P., Van Diedenhoven, B. and Zhang, Z.: Retrieval of liquid water cloud properties from POLDER-3 measurements using a neural network ensemble approach, Atmos. Meas. Tech., 12(3), 1697–1716, doi:10.5194/amt-12-1697-2019, 2019.
- Stamnes, S., Hostetler, C., Ferrare, R., Burton, S., Liu, X., Hair, J., Hu, Y., Wasilewski, A., Martin, W., van Diedenhoven, B., Chowdhary, J., Cetinić, I., Berg, L. K., Stamnes, K. and Cairns, B.: Simultaneous polarimeter retrievals of microphysical aerosol and ocean color parameters from the "MAPP" algorithm with comparison to high-spectral-resolution lidar aerosol and ocean products, Appl. Opt., 57(10), 2394, doi:10.1364/ao.57.002394, 2018.
- Xu, F., van Harten, G., Diner, D. J., Kalashnikova, O. V., Seidel, F. C., Bruegge, C. J. and Dubovik, O.: Coupled retrieval of aerosol properties and land surface reflection using the Airborne Multiangle SpectroPolarimetric Imager, J. Geophys. Res., 122(13), 7004–7026, doi:10.1002/2017JD026776, 2017.
- Xu, F., Diner, D. J., Dubovik, O. and Schechner, Y.: A correlated multi-pixel inversion approach for aerosol remote sensing, Remote Sens., 11(7), doi:10.3390/rs11070746, 2019.

# **Reviewer #3:**

## Dear Greg,

We would like to thank you for the constructive and positive comments on our paper. Please find our responses below.

This paper applies several versions of a relatively new retrieval algorithm (GRASP/HP, GRASP/Optimized, GRASP/Models) to an existing satellite measurement archive (POLDER/PARASOL). The authors then compare the results from these algorithms to several legacy retrieval algorithms, including MODIS Dark

Target (MODIS/DT), Deep Blue (MODIS/DB), Multi-Angle Implementation of Atmospheric Correction (MAIAC), the operational PARASOL product (PARASOL/Operational), and AERONET. The authors provide a large number of maps and statistics that not only inform the reader about the performance of the GRASP products, but they also inform the reader about the performance of the legacy aerosol retrievals (e.g., Tables 9 & 10). The paper is clear and well written and I find it suitable for publication.

#### **Response:**

Thank you for the positive comments on our manuscript. We provide point-by-point responses as follows.

The paper is also quite long (48 pages of text, 23 tables, and 28 figures) and probably won't be carefully read in its entirety by anyone except the reviewers. One could paraphrase the paper as "here are some new data products, and here is how they compare to similar data products as well as the gold standard (AERONET)." Nobody will learn the machinery behind the retrievals from this paper, but there are other papers for that. One reviewer pointed out that the statistical parameters chosen for this paper are not ideal, but the authors use statistical parameters that are familiar to many readers (correlation coefficient, bias, RMSE, etc.) and common in many satellite/AERONET comparison papers. Unfortunately, the aerosol remote sensing community has not yet adopted a "skill score" for comparisons, as is sometimes used in the modeling community (Taylor, 2001).

It is important to have all of this material in one place, in my opinion, so that readers can quickly assess the relative performance of the different algorithms for the various parameters. However, hyperlinks to tables, figures, citations, and section headings would greatly improve the readability of the paper. A bookmarked Table of Contents in the sidebar would also be helpful. I had to keep two copies of the paper open on my screen – one for the text, and another for reading tables and figures. Otherwise, I would have spent as much time scrolling as reading for this paper! Hyperlinks would allow the reader to go directly to a table, and then return to the text with the "previous view" buttons. Hopefully this is something that can be accommodated in the typesetting process.

Taylor, K.: Summarizing multiple aspects of model performance in a single diagram, J. Geophys. Res., 106, 7183–7192, 2001.

## **Response:**

These are very constructive suggestions. We have revised the manuscript and included a Table of Content and hyper link of all tables and figure.

I noticed that the data volume for GRASP/Models is much different than the data volume for GRASP/HP and GRASP/Optimized. For instance, GRASP/Optimized shows 41,268 AOD comparisons with AERONET over land in Table 3, but GRASP/Models only shows 27,551 comparisons. However, GRASP/Model comparisons are greater than GRASP/Optimized over ocean (2064 vs 1495). These large discrepancies appear elsewhere in the paper as well. I found this quite odd, since all three retrievals use the same instrument (PARASOL). I imagine that the cloud screening procedure is identical for all three algorithms, so I suspect that GRASP/Model fails to provide a retrieval much more frequently over land than than the other two GRASP algorithms (and that GRASP/Optimized, GRASP/HP fail more frequently than GRASP/Models over ocean). This should be discussed, since GRASP/Models is lauded for its ability to retrieve AOD (550) (e.g., line 1180). The success rate of a retrieval is important to readers, too.

## **Response:**

Indeed, the different number of points and somewhat different approaches of quality filtering is one of the main shortcomings in our study. Reviewer #1 raised similar question. In fact, the post-processing flow (L1-L2-L3) was based on several attempts dictated by practical needs. These attempts provided us valuable inside but the could be fully evaluated only after full-scale validation. For example, the level 3 GRASP/HP and Optimized archives were generated and released much earlier than GRASP/Models. Also, we have done preprocessing of GRASP/Models over land at first and learned that screening was very conservative. Based on that we used less conservative screening for Models reprocessing over ocean. Once the products were released, they were used by many users, therefore, regenerating Level 3 products was not reasonable for this study. We are considering the harmonization of the all archives in future once time and resources allow that.

At the same time, we have looked at possible effect of applying tighter screening to HP and Optimized data. Our analysis showed that although stricter screening somewhat improves the correlations, it doesn't change conceptually the results of validations. For example, it does not improve the BIAS, which is considered as a main issue for these data sets. Some explanations of this aspect were added in the Sect 2.4 and Sect 3.1 as follow.

It is also curious that GRASP/Models did so well for AOD at nearly all wavelengths over both land and ocean (Table 3), but the AEs for GRASP/Models is significantly worse than the other GRASPs. Since AE is derived from AOD, I would have thought the retrieval that produced the best AOD at multiple wavelengths would also produce the best AE. A comment about this would be helpful.

#### **Response:**

This is a good point that should be mentioned in the text. Yes, according to the analysis, we found out that the good agreement of spectral AOD is not equivalent to the good agreement for AE, even though AE is derived spectral AOD at two wavelengths. Apparently, obtaining good agreement for spectral AOD seems easier than for AE. The potential reason is that the spectral contrast is crucial to calculate AE, and small error in AOD at each wavelength can result in large AE uncertainty. The level of uncertainty (e.g.  $\pm 0.1$ , RMSE= $0.1 \sim 0.15$ ) of satellite-derived spectral AOD may makes it challenging accurate estimation of AE. At the same time, the same uncertainty is sufficient for good agreement of AOD at different wavelengths. Thus, the AOD in each channel may correlate well in time, while for each singe retrieval spectral deviations can be significant. Also, relatively small spectral deviations may perturb AE strongly while not to be as notable for AOD at each wavelength.

Is there a reason for comparing AODf and AODc to the SDA extinction-based retrievals instead of using the sky scan retrievals? The sky scans are probably more accurate. Many readers (most?) won't know the methodology behind SDA and may incorrectly assume that it is derived from the sky scans. The SDA papers use the AERONET almucantar scans as a performance benchmark, so why not use the same benchmark? You're already using the AERONET sky-scans for SSA and AAOD.

#### **Response:**

We have done AODF and AODC comparison with AERONET ALM retrievals (see in Figures R2 and R3). In general, they show quite similar performance with Figures 4 and 5 that evaluation against AERONET SDA AODF and AODC. Please note, in order to find more matched pairs, the AERONET ALM retrievals are collocated within ±180mins for satellite overpass, which is much bigger time window that that for SDA products ±30mins. To ensure the retrieval quality, AERONET ALM L2 retrieval products are available for AOD440>0.4, which roughly filter 80% low AOD cases (in future we could use AERONET L1.5 inversion products). By using SDA extinction-based products, we get almost the same amount of points for AOD, AODF, and AODC, which help to understand the overall performance for low, medium and high AOD cases.



Figure R2. Evaluation of all archive PARASOL/GRASP AODF (2005-2013) at 440 nm with AERONET INV AODF, (a) GRASP/Optimized; (b) GRASP/HP; (c) GRASP/Models.



Figure R3. The same with Figure R2, but for AODC at 440 nm

### Line 588:

The authors say that they are using AERONET L2 inversions, but which version of AERONET (i.e., Version 2 or Version 3)?

## **Response:**

We are using Version 3 AERONET L2 inversion. We now clarify this in the text.

## Lines 590-595:

This paragraph will probably confuse some people. Line 590 says that AERONET L2 inversion products require AOD(440) > 0.4, but the PARASOL/GRASP filtering includes much lower values, especially over ocean (the authors require PARASOL/GRASP AOD(443) > 0.3 over land and AOD(443) > 0.02 over ocean). However, since AERONET L2 requires AOD(440) > 0.4, many of the low PARASOL/GRASP AODs won't actually appear in the comparisons anyways. . . . Unless the authors using Level 1.5 AERONET inversions at low AOD, like some other authors? If so, what are the Level 1.5 constraints?

# **Response:**

We are using Version 3 AERONET L2 inversion products, which includes only AERONET AOD>0.4. Here we use additional filter for satellite AOD (Land: AOD 443 nm>0.3; Ocean: AOD 443 nm>0.02).

# MINOR ISSUES

Line 35:

The links do not take me directly to the data products. <u>www.icare.univ-lille.fr</u> takes me to the main page, and the 2nd link on that line tries to take me to www.grasp-35, but that is a dead end.

# **Response:**

Yes, the provided link is to ICARE main page, the PARASOL products are published at this path: <u>https://www.icare.univ-lille.fr/data-access/data-archive-access/?dir=PARASOL/</u>, ICARE account, that is free registration, is required to login. For the second link, it should work as <u>https://www.grasp-open.com/products/</u>.

Line 1125 Do you mean GRASP/Models instead of PARASOL/Models? Response: Yes, we use 'PARASOL/Models' to represent PARASOL products generated by GRASP/Models approach.

It would be interesting to repeat the AOD comparisons using AERONET's "coincident" AOD (that is, using only the AODs that are used during the sky scans). This would be interesting because the cloud screening for the sky-scan products is more comprehensive than for the direct AOD measurements, and it is possible that satellite (and model) AOD performance comparisons wrt AERONET will differ for these two datasets. If the coincident AODs comparisons are different than the "all AODs" comparisons, this could assist our thinking wrt the other sky-scan products. You are probably already set up to do this. I include this as a minor issue, though, because the paper is already too long and this should really be a topic for another day. **Response:** 

This is a good suggestion, which brings us additional thoughts! We agree that AERONET direct sun cloud screening is not as comprehensive as for sky-scan products. However, the evaluation with collocated satellite retrievals also introduces the satellite cloud screen to ensure the quality. On the other hand, when utilizing AERONET sky-scan products, in order to find more matched pairs we normally adopt a wider time window, e.g.  $\pm 180$  mins, which may increase the issue of aerosol temporal variability.

#### Validation of GRASP algorithm product from POLDER/PARASOL data assessment of multi-angular polarimetry potential for aerosol and monitoring

- Cheng Chen<sup>1, 2</sup>, Oleg Dubovik<sup>1</sup>, David Fuertes<sup>2</sup>, Pavel Litvinov<sup>2</sup>, Tatyana Lapyonok<sup>1</sup>, Anton Lopatin<sup>2</sup>, Fabrice Ducos<sup>1</sup>, Yevgeny Derimian<sup>1</sup>, Maurice Herman<sup>1</sup>, Didier Tanré<sup>1</sup>, Lorraine A. Remer<sup>3</sup>, Alexei Lyapustin<sup>4</sup>, Andrew M. Sayer<sup>4, 5</sup>, Robert C. Levy<sup>4</sup>, N. Christina Hsu<sup>4</sup>, Jacques Descloitres<sup>6</sup>, Lei Li<sup>1, 7</sup>, Benjamin Torres<sup>1</sup>, Yana Karol<sup>2</sup>, Milagros Herrera<sup>1</sup>, Marcos Herreras<sup>1</sup>, Michael Aspetsberger<sup>8</sup>, Moritz Wanzenboeck<sup>8</sup>, Lukas Bindreiter<sup>8</sup>, Daniel Marth<sup>8</sup>, Andreas Hangler<sup>8</sup> and Christian Federspiel<sup>8</sup> 5 10
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  - State Key Laboratory of Severe Weather (LASW) and Key Laboratory of Atmospheric Chemistry (LAC), Chinese Academy of Meteorological Sciences, CMA, Beijing, 100081, China Cloudflight Austria GmbH, High Performance Computing, Linz, Austria
- 20

Correspondence to: Oleg Dubovik (<u>oleg.dubovik@univ-lille.fr</u>)

Abstract. Proven by multiple theoretical and practical studies, multi-angular spectral polarimetry is ideal for comprehensive retrieval of properties of aerosols. Furthermore, a large number of advanced space polarimeters have been launched recently or planned to be deployed in the coming few years (Dubovik et

al., 2019). Nevertheless, at present, practical utilization of aerosol products from polarimetry is rather 25 limited, due to the relatively small amount of polarimetric compared to photometric observations, as well as challenges in making full use of the extensive information content available in these complex observations.

Indeed, while in recent years several new algorithms have been developed to provide enhanced aerosol retrievals from satellite polarimetry, the practical value of available aerosol products from polarimeters yet remains to be proven. In this regard, this paper presents the analysis of aerosol products obtained by the 30 Generalized Retrieval of Atmosphere and Surface Properties (GRASP) algorithm from

POLDER/PARASOL observations. After about a decade of development, GRASP has been adapted for operational processing of polarimetric satellite observations and several aerosol products from

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Cheng Chen 17/10/2020 15:36 Deleted: observations

NASA Goddard Space Flight Center, Greenbelt, MD, USA

POLDER/PARASOL observations have been released. These updated PARASOL/GRASP products are publicly available (e.g., http://www.icare.univ-lille.fr, www.grasp-open.com/products/), the dataset used in the current study is registered under: http://doi.org/10.5281/zenodo.3887265 (Chen et al., 2020).

The objective of this study is to comprehensively evaluate the GRASP aerosol products obtained 40 from POLDER/PARASOL observations. First, the validation of the entire 2005 - 2013 archive was conducted by comparing to ground-based Aerosol Robotic Network (AERONET) data. The subjects of the validation are spectral aerosol optical depth (AOD), aerosol absorption optical depth (AAOD) and single scattering albedo (SSA) at 6 wavelengths, as well as Ångström exponent (AE), fine mode AOD (AODF) 45 and coarse mode AOD (AODC) interpolated to the reference wavelength 550 nm. Second, an intercomparison of PARASOL/GRASP products with the PARASOL/Operational, MODIS Dark Target (DT), Deep Blue (DB) and Multi Angle Implementation of Atmospheric Correction (MAIAC) aerosol products for the year 2008 was performed. Over land both satellite data validations and inter-comparisons were conducted separately for different surface types, discriminated by bins of Normalized Difference Vegetation Index (NDVI): <0.2, 0.2≤ and <0.4, 0.4≤ and <0.6, and ≥0.6. Three PARASOL/GRASP 50 products were analyzed: GRASP/HP ("High Precision"), Optimized, and Models. These different products are consistent but were obtained using different assumptions in aerosol modeling with different accuracies of atmospheric radiative transfer (RT) calculations. Specifically, when using GRASP/HP or Optimized there is direct retrieval of the aerosol size distribution and spectral complex index of refraction. When using GRASP/Models, the aerosol is approximated by a mixture of several prescribed aerosol components, each 55 with their own fixed size distribution and optical properties, and only the concentrations of those components are retrieved. GRASP/HP employs the most accurate RT calculations, while GRASP/Optimized and GRASP/Models are optimized to achieve the best trade-off between accuracy and speed. In all these three options, the underlying surface reflectance is retrieved simultaneously with the aerosol properties and the radiative transfer calculations are performed "on line" during the retrieval.

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All validation results obtained for the full archive of PARASOL/GRASP products show solid quality of retrieved aerosol characteristics. The GRASP/Models retrievals, however, provided the most solid AOD products, e.g. AOD (550 nm) is unbiased, has the highest correlation (R~0.92) and the highest fraction of retrievals (~55.3%) satisfying the accuracy requirements of the Global Climate Observing

- 65 System (GCOS) when compared to AERONET observations. GRASP/HP and GRASP/Optimized AOD products show a non-negligible positive bias (~0.07) when AOD is low (<0.2). On the other hand, the detailed aerosol microphysical characteristics (AE, AODF, AODC and SSA, etc.) provided by GRASP/HP and GRASP/Optimized correlate generally better with AERONET than do the results of GRASP/Models. Overall, GRASP/HP processing demonstrates the high quality of microphysical characteristics retrieval
- 70 versus AERONET. Evidently, GRASP/Models approach is more adapted for retrieval of total AOD, while the detailed aerosol microphysical properties are limited when a mixture of aerosol models with fixed optical properties are used.

The results of a comparative analysis of PARASOL/GRASP and MODIS products showed that, based on validation against AERONET, the PARASOL/GRASP AOD (550 nm) product is of similar and sometimes of higher quality compared to the MODIS products. All AOD retrievals are more accurate and 75 in good agreement over ocean. Over land, especially over bright surfaces, the retrieval quality degrades and the differences in total AOD products increase. The detailed aerosol characteristics, such as AE, AODF and AODC from PARASOL/GRASP are generally more reliable, especially over land. The global intercomparisons of PARASOL/GRASP versus MODIS showed rather robust agreement, though some patterns and tendencies were observed. Over ocean, PARASOL/Models and MODIS/DT AOD agree well with the 80 correlation coefficient of 0.92. Over land, the correlation between PARASOL/Models and the different MODIS products is lower, ranging from 0.76 to 0.85. There is no significant global offset; though over bright surfaces MODIS products tend to show higher values compared to PARASOL/Models when AOD is low, and smaller values for moderate and high AODs. Seasonal AOD means suggest that PARASOL/GRASP products show more biomass burning aerosol loading in central Africa and dust over 85

the Taklamakan Desert, but less AOD in the northern Sahara. It is noticeable also that the correlation for the data over AERONET sites is somewhat higher, suggesting that the retrieval assumptions generally work better over AERONET sites than over the rest of the globe. One of the potential reasons may be that MODIS retrievals, in general, rely more on AERONET climatology than GRASP retrievals.

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Overall, the analysis shows that the quality of AOD retrieval from multi-angular polarimetric observations like POLDER is at least comparable to those of single-viewing MODIS-like imagers. At the same time, the multi-angular polarimetric observations provide more information on other aerosol properties (e.g. spectral AODF, AODC, AE), as well as additional parameters such as AAOD and SSA.

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	<u>1 Introduction</u>	Cheng Chen 4/10/2020 14:56
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	2.5 Considered metrics for comparison statistics	
	3 Validation of satellite observation by comparison with AERONET data: results and discussion	
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110	Aerosol Absorption Optical Depth	
	3.2 Comparison of results obtained from validation of PARASOL and MODIS aerosol products	Chang Chan 4/10/2020 14:56
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	3.3 Evaluation of PARASOL and MODIS validation results over different AERONET Sites	line: 0 cm, Line spacing: 1.5 lines
	<u>4 Inter-comparison of satellite products at global scale</u>	Chang Chan 4/10/2020 14:56
115	4.1 Comparison between PARASOL/GRASP and PARASOL/Operational aerosol products	Formatted: Indent: First line: 0 cm, Line
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#### **1** Introduction 125

Over the past few decades, satellite remote sensing has provided essential advances in understanding the global distribution of atmospheric aerosols (Kaufman et al., 2002; Remer et al., 2008) and constraining aerosol climate effects (Bellouin et al., 2005; Myhre, 2009; Yu et al., 2006). Nevertheless, aerosol effects remain the largest contributor to forcing uncertainty according to the Intergovernmental

130 Panel on Climate Change (IPCC) assessments (Boucher et al., 2013); the aerosol effective radiative forcing has been recently assessed to be between -2.0 to -0.4  $w/m_k^2$  with a 90% likelihood (Bellouin et al., 2020).

Over the past few decades, satellite remote sensing techniques have developed rapidly and extensively, and various (primarily photometric) instruments have been developed and deployed to monitor atmospheric aerosols from space (Bréon et al., 2011; Dubovik et al., 2019; King et al., 1999; Kokhanovsky et al., 2015;

- Li et al., 2009; Tanré et al., 2011). While the design and capabilites of the photometric observations are 135 constantly evolving, the greatest improvement has been in the form of Multi-Angular multi-spectral Polarimetry (MAP) measurements (Hansen et al., 1995; Hasekamp and Landgraf, 2007; Knobelspiesse et al., 2012; Mishchenko and Travis, 1997; Mishchenko et al., 2004; Waquet et al., 2009; Tanré et al., 2011). MAP measurements have enough inherent information content to greatly improve our understanding about
- 140 aerosol properties. Several space-borne polarimeters have already been deployed and more advanced versions will be deployed soon (Dubovik et al., 2019). In addition, there are many airborne versions of orbital polarimeters that have operated during field campaigns, which can be used to verify and improve the retrieval concepts (e.g. Knobelspiesse et al., 2020). Although the overall volume of polarimetric observations remains small compared to radiances-only photometric observations, the potential for rapid 145 advancement is large.

Several factors contribute to the current limited visibility of MAP observations and algorithms including: (i) limited amount of polarimetric observations in comparison to photometric ones, (ii) general complexity of polarimetric observations, and (iii) consequent challenges in developing capable retrieval algorithms. As a result, at present, there is a lack of extensive aerosol products from satellite MAPs that

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attract the aerosol science community. This tendency is especially evident by the contrast with the increase of constantly improved aerosol products from mono- and bi- viewing photometric images. For example, the archive of most popular Moderate Resolution Imaging Spectroradiometer (MODIS) observations has been processed using many different algorithms, and NASA distributes three complementary MODIS aerosol products: Dark Target (DT) by Remer et al. (2005, 2020) and Levy et al., (2013), Deep Blue by Hsu et al. (2004, 2006, 2013) and Multi Angle Implementation of Atmospheric Correction (MAIAC) by Lyapustin et al. (2018). Similarly, significant effort has been directed to improve aerosol products from European ENVISAT satellite platform observations in frame of Climate Change Initiative (CCI) projects of European Space Agency (e.g. see de Leeuw et al., 2015; Holzer-Popp et al., 2013; Popp et al., 2016). As a result, the product archives of MEdium Resolution Imaging Spectrometer (MERIS) and especially Advanced Along-Track Scanning Radiometer (AATSR) missions are constantly updated and improved.

To date, only one space-borne MAP has a long and wide enough coverage to advance aerosol science. The Polarization and Directionality of the Earth's Reflectances (POLDER) instrument was designed and developed by the French space agency Centre National d'Études Spatiales (CNES) to measure the spectral directional polarized solar radiation reflected by the Earth-atmosphere system (Deschamps et al., 1994). POLDER-1 and 2 flew on board of the Japanese Advanced Earth Observing Satellites (ADEOS) platforms ADEOS-I and JI from November 1996 till June 1997 and from April 2003 till October 2003 respectively. Unfortunately, due to the failures of the platforms' solar panels, the POLDER-1 and 2 have rather a limited time series of observations. POLDER-3 was launched in December 2004 on PARASOL (Polarization & Anisotropy of Reflectances for Atmospheric Sciences coupled with Observations from a Lidar) platform developed by CNES. POLDER-3/PARASOL (hereafter PARASOL),

was operational from March 2005 till October 2013 with nearly 5 years within the A-Train constellation, which is making nearly contiguous observations of many facets of the Earth system through a series of low-orbiting satellites (e.g. MODIS/AQUA, CALIOP/CALIPSO, OMI/AURA) (Parkinson, 2003; Schoeberl et al., 2006; Tanré et al., 2011; Winker et al., 2010). The PARASOL imager has 3 gaseous absorption

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Cheng Chen 19/10/2020 15:40 Deleted: -Cheng Chen 19/10/2020 15:40 Deleted: -Cheng Chen 17/10/2020 15:46 Deleted: correspondingly Cheng Chen 19/10/2020 15:40 Deleted: - channels (763, 765 and <u>910</u> nm), in addition to 6 channels (443, 490, 565, 670, 865 and 1020 nm) measuring the total radiance, and 3 channels (490, 670 and 865 nm) measuring the polarization. The number of viewing angles is similar for all spectral channels varying from 14 to 16 depending on the location of the pixel on the CCD, PARASOL provided global coverage about every 2 days with a nadir spatial resolution ~6 km (Tanré et al., 2011).

Several POLDER-1, 2 and PARASOL aerosol products were developed by the science team at LOA (Laboratoire d'Optique Atmosphérique, Lille, France). Hereafter, we refer to these aerosol products as 185 POLDER/Operational or Operational. The initial POLDER/Operational aerosol retrieval over ocean by Deuzé et al. (1999) provided total Aerosol Optical Depth (AOD) from the measured total and polarized radiances at 670 and 865 nm with expected accuracy of ±0.05±0.05AOD (Goloub et al., 1999). The updated algorithm by Herman et al. (2005) provided AOD of fine and coarse modes and, when geometrical 190 conditions are optimal (scattering angle ranging between  $90^{\circ}$ -160°), the spherical/non-spherical separation of coarse mode particles (Herman et al., 2005). Over land, the algorithm by Deuzé et al. (2001) retrieves only fine ("accumulation") mode AOD (AODF) using only polarized light at two wavelengths (670 and 865 nm) to capitalize on the small and fairly neutral polarized reflectance typical of land surfaces (Deuzé et al., 2001; Herman et al., 1997). These algorithms were designed to utilize the benefits of MAP information within the framework of a conventional MODIS like Look-up-Table (LUT) approach (Tanré et al., 1997; 195 Kaufman et al. 1997) and did not intend to extend substantially the set of retrieved parameters. Moreover, over land the POLDER/Operational retrieval provided only AODF, more sensitive to fine mode, while MODIS algorithms derives the total AOD.

The Generalized Retrieval of Atmosphere and Surface Properties (GRASP) algorithm considered here was developed to further exploit the aerosol information content of POLDER spectral multi-angular polarization measurements (Dubovik et al., 2011, 2014). The algorithm allows for a large number of unknown parameters and retrieves a set of parameters affecting measurements at all wavelengths, all angles, and all states of polarization using the multi-term least square method (Dubovik, 2004). As will be later

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Utilisateur Microsoft O..., 2/11/2020 12:59 Formatted: English (US) Cheng Chen 17/10/2020 15:49 Deleted: this described in detail in section 2.1, GRASP does not utilize pre-calculated LUTs but instead searches in a

- 210 continuous space for the solutions and optimizes the statistical properties of the obtained retrieval. The GRASP algorithm derives an extended set of aerosol parameters from POLDER data, including spectral AOD, spectral Aerosol Absorption Optical Depth (AAOD), spectral AODF, spectral AODC, particle size distribution, Single Scattering Albedo (SSA), complex refractive index, fraction of spherical particles, etc. (see Table 1 and discussion in the next Section). The full archives of POLDER-1, 2 and PARASOL were
- 215 processed with GRASP and the resulting datasets are available for public at the official GRASP algorithm website (<u>www.grasp-open.com</u>) and the AERIS/ICARE Data and Services Center (<u>http://www.icare.univ-lille.fr</u>).

This paper presents and discusses new publicly-available aerosol products generated by the recently developed GRASP algorithm (Dubovik et al., 2011, 2014) applied to PARASOL observations, which 220 represent the longest to date satellite MAP record (Tanré et al., 2011; Dubovik et al., 2019). Hereinafter we perform quantitative analysis of PARASOL/GRASP aerosol products (the longest POLDER data set) through validation with AERONET reference data, as well as by comparisons with the operational products and the widely used MODIS DT, DB and MAIAC aerosol products. The analysis pursues two objectives. The first is to understand the accuracy and value of each PARASOL/GRASP aerosol product. The second objective is to clarify the specifics, advantages, and shortcomings of MAP aerosol products compared to 225 those from radiances-only photometric mono-viewing imagers. Thus, the analysis provides useful information for the aerosol community to meet the future challenge of accurate aerosol monitoring in the coming era of polarimetric missions. Over the next few years, we expect deployment of a number of new and existing satellite and airborne MAPs including 3MI (Multi-View Multi-Channel Multi-Polarization Imaging), DPC (Directional Polarimetric Camera), Aerosol-UA (Ukraine), PACE (Plankton, Aerosol, 230 Cloud, ocean Ecosystem), AirHARP (Airborne Hyper-Angular Rainbow Polarimeter), AirMSPI (Airborne Multi-angle SpectroPolarimeter Imager), SPEXone (Spectro-Polarimetric Experiment), RSP (Research Scanning Polarimeter), etc. (Dubovik et al., 2019; Fougnie et al., 2018; Fu et al., 2020; Gao et al., 2020;

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Hasekamp et al., 2019a; Knobelspiesse et al., 2020; Li et al., 2018; Milinevsky et al., 2019; Puthukkudy et al., 2020; Remer et al., 2019), Several studies have shown the potential of advanced MAP aerosol products, for example, PARASOL/GRASP results have been adopted to estimate global aerosol emissions (Chen et al., 2018; 2019), PARASOL/SRON products have been used for data assimilation (Tsikerdekis et al., 2020), estimation of aerosol direct radiative effect (Lacagnina et al., 2015; 2017), and the radiative forcing due to aerosol-cloud interactions (Hasekamp et al., 2019b). By providing a comprehensive analysis of PARASOL/GRASP products, we intend to prove that the aerosol community can utilize the new era MAP

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2 Data description and validation approach

measurements.

The analysis compares several satellite data products. From POLDER, we have both the products of the Operational algorithm and the GRASP retrieval. From MODIS, we utilize products generated by three different algorithms (DT, DB, and MAIAC). For all satellite products, validation is based on AERONET observations and retrievals.

#### 2.1 POLDER/GRASP aerosol products

GRASP is a new-generation algorithm developed for deriving extensive aerosol properties from all remote sensing instruments. The overall concept of the algorithm is described by Dubovik et al. (2014), while specific technical aspects are detailed in Dubovik et al. (2011). GRASP is based on highly advanced statistically optimized fitting implemented as multi-term least square minimization (Dubovik, 2004) which had earlier been successfully implemented for aerosol retrievals from ground-based AERONET radiometers (Dubovik and King, 2000; Dubovik et al., 2000, 2002a, 2002b, 2006). GRASP inherits many methodological aspects in numerical inversion and aerosol modeling from the AERONET retrieval developments. In fact, all retrieval set-ups including modeling of aerosol microphysical and optical properties, surface reflectance, numerical inversion, utilization of multiple a priori constraints, etc. can be realized using GRASP. At the same time, the GRASP concept and algorithm are highly flexible and

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versatile. GRASP includes several additional original features, and enables the implementation of advanced retrieval scenarios. A unique aspect of GRASP is that it can perform radiative transfer (RT) computations fully accounting for multiple interactions of the scattered solar light in the atmosphere <u>online without the</u> use of traditional LUTs, Several other algorithms of new generation have been or being developed for

interpretation of MAP observation use the online RT calculations and implement retrieval as a search in continuous space of solution e.g. Hasekamp et al., 2011, Xu et al., 2017, 2019, Fu and Hasekamp, 2018, Gao et al., 2018, Stamnes et al., 2018, Di Noia et al., 2019, Nonetheless, at present GRASP was the only algorithm that has been used to generate aerosol products for full archive of POLDER observations. (Dubovik et al., 2019).

The GRASP retrieval can utilize whatever information content is available. If there is sufficient information content of the observations, GRASP will find the aerosol solutions. In the case of any currently operational observations, GRASP can make optimal assumptions to constrain the solution. For example, GRASP can retrieve both aerosol and underlying surface properties simultaneously from <u>multispectral</u> satellite observations using additional a priori constraints on the spectral variability of land Bidirectional

Reflection Distribution Function (BRDF). Or (probably the most essential methodological novelty) it can operate by relying on the multi-pixel concept wherein the statistically optimized retrieval is performed simultaneously for a large group of pixels (Dubovik et al., 2011). This feature brings additional possibilities for improving the accuracy of satellite retrievals by using known constraints on the inter-pixel variability of retrieved aerosol and surface reflectance parameters. As a result, using this methodology GRASP provides reliable retrievals of detailed aerosol properties that traditionally have been difficult to obtain from satellites, for example, spectral AOD and AAOD over land including very bright deserts. The GRASP algorithm source code and detailed documentation are available from https://www.grasp-open.com.

It should be noted that GRASP is a flexible inversion algorithm that can be applied to a wide variety of satellite, ground-based and laboratory observations. It has already been applied to ground-based 295 AERONET photometers and LiDARs (Benavent-Oltra et al., 2017, 2019; Hu et al., 2019; Lopatin et al.,

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2013; Titos et al., 2019; Tsekeri et al., 2017), sky cameras (Román et al., 2017), polar-nephelometer data (Espinosa et al., 2017, 2019; Schuster et al., 2019), and surface measurements of AOD (Torres et al., 2017).

- 315 In addition, GRASP is being used for several satellite instruments; aerosol products were generated for POLDER observations (discussed here) and for MERIS/Envisat, and there are ongoing developments for producing GRASP aerosol products from Sentinel-3 and -5P observations and operational aerosol retrievals for future Sentinel-4 and 3MI/Metop missions. GRASP is constantly being updated to produce many useroriented products such as estimates of covariance matrices (Herrera et al., in preparation, 2020), direct
- 320 radiative forcing (Derimian et al., 2016), and so on.

For POLDER, GRASP utilizes radiance and polarization observations from all available spectral channels, including minor gaseous absorption for some of them, i.e. for total radiance 5 channels for POLDER-1 and 2, and 6 for PARASOL, and for polarized radiances (3 spectral channels for all instruments). The retrieval uses a unique global set of constraints (no location-specific assumptions) and a

325 single initial guess globally. GRASP performs radiative transfer computations fully accounting for multiple interactions of the scattered solar light in the atmosphere online without using a traditional LUT. Since these RT computations are complex and time consuming, significant effort has been put into optimization and acceleration of the code for operational processing of voluminous datasets. At present, the speed of GRASP retrieval is appropriate for processing the full archive of POLDER observations at native 330 resolution (POLDER-1 and 2 at ~7 km and PARASOL at ~6 km) using rather moderate computing

resources, e.g. 3-4 sec/pixel for GRASP/HP, 0.3-0.5 sec/pixel for GRASP/Optimized and 0.1-0.2 sec/pixel for GRASP/Models, in a single core processor (the description of GRASP/HP, GRASP/Optimized and GRASP/Models will be detailed further in this section).

Since GRASP has been designed for use with different observations, it allows a variety of different 335 possibilities on modeling aerosol scattering, surface reflectance and generally on implementing atmospheric radiative transfer calculations. As a result, different configurations of the atmospheric forward model can be used even for interpretation of the same data (as is the case here with POLDER). Currently,

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1) PARASOL/GRASP «optimized» (in the sense that radiative transfer calculations were optimized to

find the best trade-off between speed of processing and accuracy of results);

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- PARASOL/GRASP «high-precision» (radiative transfer calculations with high precision were used).
- PARASOL/GRASP «models» (the simplest, fastest processing; aerosol is assumed to be external mixture of several aerosol models).
- The «optimized» and «high-precision» are different only by the <u>online</u> precision of the RT calculations, while they are <u>conceptually</u> the same: aerosol size distribution, spectral values of complex index of refraction, fraction of spherical particles and the Aerosol Layer Height (ALH), are retrieved simultaneously with the surface BRDF and Bidirectional Polarization Distribution Function (BPDF) parameters. The retrievals were performed using one aerosol component model with 5 bins of the size distribution and spectrally dependent complex refractive index. The aerosol vertical distribution was modeled using an exponential profile and scale height was retrieved. The details of implementation are discussed in <u>Dubovik</u>
- et al. (2011). The «models» approach uses different assumption for modeling aerosol properties (surface treatment is the same as above): aerosol is assumed to be an external mixture of several aerosol components and only their<u>respective</u> concentrations are retrieved together with ALH and spectral BRDF/BPDF parameters. The size distribution, complex refractive index and non-sphericity parameter for each aerosol component are derived from the results of AERONET aerosol climatology for the main distinct aerosol types (Dubovik et al., 2002b) and improved in a series of sensitivity tests with satellite data. For retrievals over land, GRASP retrieves the parameters of the Ross-Li BRDF (Li and Strahler, 1992; Ross, 1981) and BPDF (Maignan et al., 2009) models under assumption that the retrieved parameters are spectrally smooth (the strength of smoothness is different for each parameter) (Litvinov et al., 2011a, 2011b). For retrievals over ocean, the wind speed and a spectrally dependent Lambertian albedo are included in the state vector. It

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370 observations such as those from MERIS/Envisat. However, once the approach was tested with PARASOL data, the obtained results were quite appealing especially in conditions of low aerosol loading, motivating the generation of PARASOL/GRASP «models» archive that is included in the consideration here.

The three archives (Optimized, HP and Models) are released publicly and can be found at the AERIS/ICARE Data and Services Center (<u>http://www.icare.univ-lille.fr</u>) and at GRASP-OPEN website

(https://www.grasp-open.com/products/) in slightly different formats. The AERIS/ICARE is the official distributor of POLDER Level-1 and 2 data and allows the user to dive into the data using a web tool, which plots the results online. The AERIS/ICARE provides detailed visualization of the data, while GRASP-OPEN site is faster in releasing new products but with no visualization. The original PARASOL/GRASP retrievals are stored at Level-1, Level-2 and Level-3 products and are publicly available in the form of daily, monthly, seasonal, yearly and climatological datasets. The Level-2 data contain full resolution data filtered following established quality criteria. Level 3 data is aggregated into a 0.1° and 1° grid box using the sinusoidal projection and gdalwarp regridding technique (https://gdal.org/programs/gdalwarp.html) from arithmetic mean of Level-2 data. As discussed in Sayer and Knobelspiesse (2019), the arithmetic vs. geometric differences are likely significantly smaller for Level 3 0.1° data used in this study. The list of

retrieved aerosol parameters, as well as derived aerosol characteristics can be found in the Table 1. In this study, we adopt the current latest version of Optimized, HP (v1.2) and Models (v2.1) products.

## [Table 1]

In addition to the PARASOL/GRASP products, all observations of POLDER-1 and 2 were also processed (using the GRASP/Models approach only). These data records are much shorter than PARASOL and therefore not included in the following analysis. However, based on limited comparisons (not presented here), the quality of the POLDER-1 and 2/GRASP retrievals is expected to be similar to those of PARASOL/GRASP retrievals. Also, recently a new "GRASP/Component" approach has been developed (Li et al., 2019, 2020a, 2020b). This approach retrieved the size resolved fractions of aerosol components Cheng Chen 19/10/2020 16:16 Deleted: -

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representing the different composition species, like black carbon, brown carbon, fine/coarse mode nonabsorbing soluble and insoluble, coarse mode absorbing and aerosol water. The retrieved fractions drive the aerosol spectral index of refraction in modeling atmospheric radiances. This provides a fourth retrieval archive; however, the results have not yet been fully analyzed and are not released in a user friendly format, so the GRASP/Component data set will not be considered in this study.

PARASOL/GRASP aerosol products have already appeared in many studies, i.e. validation (Tan et al., 2019; Wei et al., 2019, 2020), data assimilation (Chen et al., 2018, 2019), AOD products merging (Li et al., 2020; Sogacheva et al., 2020). Despite these preliminary applications of the products, no systematic evaluation of the global PARASOL/GRASP aerosol products has been published. Moreover, most early studies are based on the GRASP/Optimized products, which were released first. The evaluation of PARASOL/GRASP surface properties, as well as aerosol microphysical parameters (size distribution, complex refractive indices, fraction of spherical particles), and aerosol layer height, will be the subject of separate studies.

#### 2.2 MODIS Dark Target, Deep Blue and MAIAC aerosol products

The MODIS sensors on board TERRA since 2000 (overpass ~10:30 local) and AQUA since 2002 (overpass ~13:30 local like PARASOL during the 5 first years) provide near-global coverage twice per day. In this study, we will employ products from MODIS/AQUA only, which is on the same A-Train afternoon constellation orbit as PARASOL. MODIS has a wider swath of 2330 km compared to ~1600 km of PARASOL, 36 spectral channels ranging from 410 to 15000 nm and higher spatial resolution for cloud mask. There are 3 mature aerosol products produced operationally and distributed by NASA: *Dark Target*,

Deep Blue and MAIAC.

420 MODIS Dark Target

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The Dark Target (DT) algorithm over land is based on an empirical surface reflectance relationship between blue and red channels with the shortwave infrared (2113 nm) radiance. The AOD is retrieved by matching LUT values to observations at 466 nm, and then varying the weighting between two fixed aerosol models until the residual between LUT and observations are minimized at 645 nm. The main product is

- 425 AOD at 553 nm with AOD reported at 466 nm, 645 nm and 2113 nm, consistent with the selected weighted aerosol model (Kaufman et al., 1997; Levy et al., 2007a, 2007b). Over ocean, the simplicity of the dark ocean surface permits the retrieval of AOD and aerosol particle size (Tanré et al., 1997). In this situation the algorithm chooses one fine mode out of four and one coarse mode out of five, along with the relative weight between fine and coarse mode by minimizing the summed difference between LUT and
- observations in six wavelengths (550, 660, 870, 1240, 1610, and 2130 nm) (Tanré et al., 1997; Remer et al., 2005, 2020; Levy et al., 2013). The MODIS DT aerosol products are periodically updated to improve overall performance (Levy et al., 2003, 2007a, 2007b, 2013; Remer et al., 2005; Gupta et al. 2016). The widely recognized limitation of the DT algorithm is the complex spectral structure of bright land surfaces (e.g. desert, bare soil, snow) that violates the assumptions of the empirical relationships between
  wavelengths and increases uncertainty in the aerosol retrievals to unacceptable levels. Therefore, DT does not provide coverage over these cases.

## MODIS Deep Blue

The Deep Blue (DB) algorithm retrieves over both bright (except snow) and vegetated land surfaces. It is able to retrieve over brighter surfaces than DT because it makes use of the much darker surface reflectance in the deep blue (412 nm) channel (Hsu et al., 2004, 2006, 2013). Depending on the processing path, determined by observed reflectance and vegetation indices, the algorithm will invoke empirical spectral relationships of surface reflectance similar to DT (vegetation), rely on a pre-calculated data base of surface reflectance (arid/deserts) or apply a hybrid method (urban surfaces). The MODIS DB aerosol products have also gone through several version updates (Hsu et al., 2013; Sayer et al., 2015).

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Within the MODIS official products, the DB algorithm is applied for only land aerosol retrieval. Over

vegetated surfaces DT tends to provide more retrievals in the tropics, and DB more retrievals at midlatitudes, due to different pixel selection and cloud screening criteria (Sayer et al., 2014).

#### 450 MODIS MAIAC

The Multi Angle Implementation of Atmospheric Correction (MAIAC) algorithm has been developed and applied to MODIS (Lyapustin et al., 2011a, 2011b, 2012, 2018), and is running operationally in the NASA system. The MAIAC algorithm uses the minimum reflectance method to dynamically characterize spectral ratios of the surface reflectance (which are prescribed in the DT) and separate aerosol and surface contributions to the measurements. The accumulation of up to 16 days of the last observations in the operational memory allows MAIAC to derive spectral surface BRDF. The MAIAC aerosol product is available at higher spatial resolution of 1 km, in comparison to DT and DB that provide aerosol products at 3 km and 10 km. As a more recent addition to the MODIS family of aerosol products than DT and DB, MAIAC has shown itself to produce an AOD product as accurate or better than the older algorithms over all types of land surfaces (Jethva et al., 2019), and thus offers a complementary/alternative product to those from the original DT and DB algorithms.

All three MODIS algorithms (DT, DB and MAIAC) are developed based on LUT approaches with a fixed certain number of aerosol models. Over ocean, DT assumes 9 aerosol models (4 fine models plus 5 coarse models); any retrieval corresponds to one of total 20 combinations of one fine mode and one coarse mode (Levy et al., 2003; Remer et al., 2005; Tanré et al., 1997). Over land, DT algorithm adopts aerosol models from AERONET retrievals, clustering down to three possible spherical fine-mode dominant models (non-absorbing, moderately-absorbing and absorbing) and 1 spheroid coarse-mode dominant model (Levy et al., 2007a). In addition, the fine- and coarse-mode dominant aerosol models over land are defined as a function of season and location (Levy et al., 2013). The DB algorithm makes use of prescribed dust, and smoke/sulfate aerosol models in the LUT (Hsu et al., 2013). For example, over vegetated surfaces, AE is

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limited to some extent  $(0.0 \le AE \le 1.8)$ , and fixed at 1.5 for low AOD conditions. Over bright arid/desert

surfaces the AE is limited to a maximum of 1.0 (Hsu et al., 2013; Sayer et al., 2013). A geographic distribution of aerosol models is also adopted in the MAIAC algorithm, where the aerosol model parameters are regionally, and may be parameterized as a function of AOD (dynamic models) for regions

- 475 with high humidity variations. The detailed description of the MAIAC regional aerosol models can be found in Lyapustin et al., (2018). Hence, the MODIS aerosol products do not have the ability to retrieve aerosol particle properties with known uncertainties, with the exceptions of size parameter (over ocean in DT), SSA for dust (in DB), and AE (with known caveats).
- In this study, MODIS Collection 6 aerosol products (MYD04\_L2) from DT and DB algorithms were acquired from the AERIS/ICARE Data and Services Center (http://www.icare.univ-lille.fr, last access: 30 August 2019), where the unchanged NASA MODIS data are redistributed with enhanced visualization. Note that the latest versions of DB and DT are Collection 6.1, although the differences between the two versions are small on a large scale (Sayer et al., 2019) and do not significantly affect the conclusions presented here. The latest MAIAC Collection 6 aerosol data (MAC19A2) is obtained from NASA LAADS (Level-1 and Atmosphere Archive and Distribution System) DAAC (Distributed Active
- Archive Center) (https://ladsweb.modaps.eosdis.nasa.gov, last access: 8 January 2020).

## **2.3 AERONET Dataset**

The Aerosol Robotic Network (AERONET) is a global distributed network of well-calibrated sunsky photometers (Holben et al., 1998). By measuring direct Sun radiance, AERONET provides high temporal (every 3 or 15 minutes in daytime depending on the operation mode of the instruments) multiwavelength AOD products with high reliable accuracy (~±0.01 to ±0.02) (Eck et al., 1999). Strict protocols for the calibration and maintenance assure homogeneity among all its instruments. Due to its high data quality, the AERONET AOD products are widely used as "ground truth" to evaluate satellite remote sensing aerosol products (Bréon et al., 2011; Chu et al., 2002; Kahn et al., 2005; Liu et al., 2004; Remer et 495 al., 2005, 2002; Sayer et al., 2013). In addition to direct Sun observations, AERONET radiometers conduct routine measurements of the sky-scanning diffuse radiation. These observations are used to derive aerosol microphysical properties, e.g. single scattering albedo, complex refractive index, size distribution and sphericity via Dubovik and King (2000). The accuracy of the AERONET inversion products has been analyzed in many studies (Dubovik et

al., 2000; Sinyuk et al., 2020) and resulting recommendations were adopted for providing aerosol products of highest quality (e.g. increase of quality of retrieval products with aerosol loading and range of observed scattering angles). The microphysical properties provided by AERONET contribute to aerosol and climatic applications. For example, the AERONET-derived aerosol particle property climatology (Dubovik et al., 2002b), are used in some form in nearly all satellite retrieval algorithms (including MODIS, see Levy et al.,

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<sup>55</sup> 2007b; Lyapustin et al., 2018) and feed the climate models used to characterize aerosol climate effects (Kinne et al., 2003; Sato et al., 2003).

In this study, the up-to-date AERONET Version 3 Level 2.0dataset (http://www.aeronet.gsfc.nasa.gov, last access: 3 September 2019) (Giles et al., 2019) with standard cloud screening and quality control were used (Smirnov et al., 2000). We make use of all AERONET sites with data during the POLDER/PARASOL archive (2005-2013). The AERONET direct-sun AOD, Ångström 510 Exponent, fine and coarse mode AOD from spectral deconvolution algorithm (SDA) (O'Neill et al., 2003), AAOD and SSA products are chosen as references for satellite products comparison and evaluation.

### 2.4 Data quality assurance and matchup methodology

One of the main issues in satellite data validation is how to match the temporally-varying 515 AERONET point measurements with the spatially-varying satellite remote sensing aerosol products at over pass time (Ichoku et al. 2002). This issue is compounded when multiple satellite products are involved that vary from ~1 km to ~100 km pixel spatial resolution. There are some insightful studies (Kinne et al., 2013; Schutgens et al., 2017) that quantify the AERONET sites spatial representativeness at the scales from ~50 km to ~300 km, which can be used for evaluation of chemical transport model simulations. However, the

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Cheng Chen 19/10/2020 16:28 Deleted: in spatial resolutions (~50 km to ~300 km) considered in those studies are seemingly too coarse for validation of satellite products of 1 km for MAIAC, 10 km for DB and DT and ~6 km data from PARASOL/GRASP.

This study considers aerosol products at 10 km spatial resolution; that is the native resolution of MODIS DB and DT products and seems to be the best compromise for comparing PARASOL/GRASP,

MODIS DT, DB and MAIAC results. Also, 10 km is utilized by the aerosol community and other datasets 525 (e.g. ESA CCI products mentioned earlier). That also was a reason for the generation of PARASOL/GRASP Level 3 products. Thus, we adopted PARASOL/GRASP Level 3 daily 0.1° gridded aerosol products, MODIS/AQUA Level 2 daily DT and DB 10 km products and the 1 km MODIS MAIAC aggregated to 0.1° (MAIAC 0.1) and 0.01° (MAIAC 0.01) resolution for the inter-comparisons. MAIAC 0.01 essentially represents the single 1 km pixel retrieval. The PARASOL/Operational L2 daily 530 aerosol product is directly used for validation, which is at 18.5 km x 18.5 km spatial resolution.

The strategies to select PARASOL/GRASP retrieval products with highest quality are presented in Table 2. The land pixel is defined only if 100% of the  $0.1^{\circ}$  by  $0.1^{\circ}$  grid box has been identified as land, so an ocean pixel must contain 0% land. Also, to guarantee proper coast elimination, the first pixel bordering ocean and land is removed (see Fig. 1). We selected the more reliable retrievals using "Residual Relative" 535 (mean-root-square of relative error in fitting the measurements by the algorithm) for PARASOL/GRASP products. We adopted the same threshold for GRASP/Optimized and GRASP/HP (0.05 over land and 0.1 over ocean). These thresholds are suggested for general users. For GRASP/Models product we did not use any filter because a stricter quality assurance filter has been applied in GRASP/Models products generation from L1 to L2 and L3 than for other GRASP datasets. In principle, the post-processing of all 540 PARASOL/GRASP products was done in similar ways. At the same time, the L3 products were prepared and released not at the same time. For example, the L3 GRASP/HP and Optimized archives were generated and released much earlier than GRASP/Models, Therefore, the post-processing and quality screening approaches used for different data archives are not exactly the same. Unfortunately, most of the differences

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data quality filter. While a general filter is suggested for GRASP/Optimized and GRASP/HP products analysis, since the filter for products generation is

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not as strict as for GRASP/Models. Chena Chen 2/11/2020 22:0

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were identified after the release of the products, its extensive use and the full-scale validation. In these

regards, the harmonization of the all archives is likely to be done in future, but it will lead in release of new

products 570

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In this study we tried to avoid additional filtering of PARASOL/GRASP L3 products, since most of users utilize the products with no screening or with a very straightforward filtering. For MODIS DT, DB and MAIAC products, we select the data only with the highest Quality Assurance (QA) flag. The highest "Quality Index" was selected for PARASOL/Operational products (Bréon et al., 2011). Any pixel with fitting residual higher than the threshold for PARASOL or QA lower than the highest flag for MODIS will be set to "no data".

## [Table 2]

For validation with AERONET over land, we averaged all land satellite retrievals in a 3x3 window for the gridded satellite data centred over the AERONET station. For ocean sites, in order to select pure 580 ocean pixels and keep reasonably high number of validation points, we decided to use a 9x9 window over the AERONET site, using only pure ocean pixels. Any ocean pixels adjacent to land or land-ocean mixed pixels were omitted as represented in Figure 1. The minimal number of accepted satellite data pixels within the window is 1 over land and 41 over ocean; otherwise, the data were excluded from comparison. The PARASOL/Operational product is treated a bit differently over ocean due to its relatively coarse resolution (~18.5 km), with a similar land-like 3x3 window centred over the AERONET station. 585

The AERONET direct-sun AOD, AE, AODF and AODC data were averaged within ±30 minutes of the MODIS/AQUA and PARASOL overpass time, while AERONET SSA and AAOD (which have a lower sampling frequency) are averaged within ±180 minutes. In addition, AERONET station elevations greater than 3600 m above mean sea level, satellite 3x3 or 9x9 data sets with AOD standard deviation greater than 0.05 between window pixels were excluded.

[Figure 1]

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#### 2.5 Considered metrics for comparison statistics

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For quantifying the validation results, we used standard statistical parameters, including Pearson's linear correlation coefficient (R), root mean square error (RMSE), slope and offset of linear regression and bias.

$$R = \frac{\sum_{i=1}^{N} (O_{i,satellite} - O_{satellite}) (O_{i,AERONET} - O_{AERONET})}{\sqrt{\sum_{i=1}^{N} (O_{i,satellite} - \overline{O}_{satellite})^2 \sum_{i=1}^{N} (O_{i,AERONET} - \overline{O}_{AERONET})^2}}$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^{N} (O_{i,satellite} - O_{i,AERONET})^2}{N}}$$
(2)

$$BIAS = \frac{1}{N} \sum_{i=1}^{N} (O_{i,satellite} - O_{i,AERONET})$$
(3)

where *N* is the number of matched data points *i*;  $O_{\text{satellite}}$  represents the observations from satellite, and  $O_{\text{AERONET}}$  represents the referenced observations from AERONET;  $\overline{O_{\text{satellite}}}$  and  $\overline{O_{\text{AERONET}}}$  are the mean value for satellite and AERONET observations.

- For MODIS validation, a commonly-used metric is the fraction agreeing within and Expected Error (EE) envelope such as  $\pm 0.05\pm 0.15$ AOD (Remer et al., 2005) or  $\pm 0.05\pm 0.1$ AOD (Lyapustin et al., 2018). In this study, we adopted stricter requirements proposed by the Global Climate Observing System (GCOS) (the greater of 0.03 or 10%), which have been adopted in the Aerosol\_cci study (Popp et al., 2016) as well as the latest DB validation (Sayer et al., 2019). Following the Aerosol\_cci study by Popp et al. (2016), the uncertainty of 0.01 for AERONET AOD has been taken into account and GCOS is defined as: GCOS = max( $\pm 0.04, \pm 0.1$ AOD) (4)
- 625 Hence, the GCOS fraction (%) is the percentage of satellite retrieved AOD satisfying the GCOS requirement.

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### 3 Validation of satellite observation by comparison with AERONET data: results and discussion

- In order to characterize the quality of the retrieved aerosol parameters from PARASOL, the set of main aerosol parameters including AOD, AE, AODF, AODC, SSA and AAOD were evaluated for the 755 entire PARASOL ~9 years (2005-2013) data archive. This list includes all main aerosol parameters expected to be retrieved from MAP instruments in general (Dubovik et al., 2019). In addition, the validation results of AOD, AE, AODF and AODC were compared with the results of validation of these (where available) from the standard PARASOL/Operational and MODIS products for the year 2008.
- PARASOL/GRASP retrievals are available and validated at six wavelengths (443, 490, 565, 670, 760 865 and 1020 nm). The MODIS retrievals and even PARASOL/Operational have different spectral coverage and, therefore, the comparisons of the GRASP product focused on the aerosol properties at midvisible (550 nm) that is commonly used in the satellite data comparisons and analysis (e.g. Saver et al., 2018; Sogacheva et al., 2020). Therefore, for PARASOL/GRASP and PARASOL/Operational data the aerosol products were generated at 550 nm by interpolations in log-log space from the closest channels 765 available from the products. Similarly, AERONET aerosol products were also interpolated to 550 nm since the ground-based radiometers do not have a 550 nm channel.

#### 3.1 Global validation of PARASOL/GRASP aerosol products

#### Aerosol Optical Depth

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Fig. 2 shows scatter plots of co-located PARASOL/GRASP AOD against AERONET AOD at 550 nm for the entire POLDER/PARASOL archive; Fig. 2a for GRASP/Optimized; Fig. 2b for GRASP/HP; and Fig. 2c for GRASP/Models. Validation metrics for total spectral AOD (443, 490, 550, 565, 670, 865 and 1020 nm), as well as AOD separated for land and ocean, are presented in Table 3. As can be seen from Fig. 2 and Table 3, all retrievals present good agreement with AERONET spectrally. Overall, based on these metrics the quality of the comparison with AERONET is best for GRASP/Models. For example, for 775 AOD (550 nm) GRASP/Models shows better performance than GRASP/Optimized and GRASP/HP: R=0.923 as compared to 0.877 and 0.899 and RMSE=0.119 for GRASP/Models as compared to 0.160 and

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0.161 for GRASP/Optimized and GRASP/HP respectively at 550 nm (see in Fig. 2). GRASP/Optimized 780 and GRASP/HP show a positive overall bias of 0.06-0.07 for all AOD conditions, that remains for low AOD conditions (AOD<0.2) and even increases to 0.08 (GRASP/Optimized). In comparison, GRASP/Models has small overall bias (of 0.01 for AOD at 550 nm) that slightly increases to 0.03 for high AOD conditions (AOD>0.7). Because of the bias in GRASP/HP and GRASP/Optimized AOD, GCOS fraction for them is much lower than for GRASP/Models AOD: e.g. 55.3% (AOD at 550 nm) for land + 785 ocean vs. 28.2% and 34.4% respectively. Over ocean, all three archives show good correlation with coefficients R > 0.93 at 550 nm. Nevertheless, GRASP/Models over ocean has the highest R = 0.950 and offers the best performance for the other statistical metrics. As described in section 2.4, the different postprocessing scheme resulted in the difference for matched points between GRASP/Optimized, GRASP/HP 790 and GRASP/Models. It can be noticed that applying a much stricter filter may improve the overall correlation against AERONET AOD for GRASP/HP and GRASP/Optimized products, but leads to significant loss of points and, most importantly, do not improve the BIAS, which is considered as a main issue for them.

It is very important to note the robust performance of PARASOL/GRASP AOD retrieval in all spectral channels. For example, GRASP/Models product shows only minor spectrally independent bias of 0.01 over land, and over ocean the bias is about 0.02 at 440 nm and decreases to zero at longer wavelengths, and the GCOS fraction for all wavelengths is at least ~50% over land and ~60% over ocean.

## [Figure 2]

[Table 3]

800 Ångström Exponent

AE was determined from AOD at two different wavelengths (AE =  $\frac{\ln [\tau(\lambda_1)/\tau(\lambda_2)]}{\ln (\lambda_2/\lambda_1)}$ ). The accuracy of

AE decreases for low AOD because even a small spectral bias the AOD affects AE strongly (e.g., Wagner and Silva, 2008). Therefore, the threshold of PARASOL AOD (550 nm) > 0.2 was used in AE validation. For calculating the PARASOL AE (440/870), the AOD retrieved at 443 nm and 865 nm are interpolated to Cheng Chen 8/11/2020 12:35 Formatted: (Asian) Chinese (PRC) Cheng Chen 17/10/2020 16:25 Deleted: 4 Cheng Chen 8/11/2020 12:35 Formatted: (Asian) Chinese (PRC) Cheng Chen 8/11/2020 12:35 Formatted: Caption, Centered, Line spacing: single Cheng Chen 4/10/2020 14:49 Formatted: Font:Not Bold Cheng Chen 4/10/2020 14:48 Formatted: Heading 3, Line spacing: Cheng Chen 4/10/2020 14:48 Deleted: Angström Exponent -



nominal 440 nm and 870 nm wavelengths. Fig. 3 shows the scatter plots of PARASOL/GRASP AE against AERONET AE (440/870) for the whole archive (Fig. 3a: for GRASP/Optimized, Fig. 3b: for GRASP/HP and Fig. 3c: for GRASP/Models). GRASP/HP has a higher correlation R (0.845) than GRASP/Optimized

810 (0.800) and GRASP/Models (0.692). In addition, GRASP/HP shows a lower RMSE (0.334) than GRASP/Optimized (0.356) and GRASP/Models (0.415). The statistics of separated land and ocean AE validation are presented in Table 4. Over ocean, the correlation coefficients are significantly higher (R>0.93) than over land for all three datasets. Overall, the AE correlation statistical metrics is the best for GRASP/HP both over land and ocean. GRASP/Models product has the smallest BIAS over land, which is
815 counterpoised by overestimation of low and underestimation of high AE values due to assumed size distributions in the agraent model bacad approach. Both GRASP/Optimized and GRASP/HP approach.

distributions in the aerosol model-based approach. Both GRASP/Optimized and GRASP/HP capture AE well when large particles are dominant (AE<1.0), while the products tend to slightly underestimate AE when small particles are dominant (AE>1.0).

#### 820

# [Figure 3] [Table 4]

## Fine- and Coarse- mode Aerosol Optical Depth

Fig. 4 shows the validation of PARASOL/GRASP AODF against SDA AODF provided by AERONET. AERONET SDA products (O'Neill et al, 2003) reported only at 500 nm, therefore here were interpolated to AODF at 550 nm based on AE using a quadratic fit in log-log space (Eck et al., 1999). Over land + ocean, GRASP/HP AODF shows the best validation statistics with correlation R=0.925, BIAS=0.01 and Slope=0.892 compared to R=0.922, BIAS=0.02 and Slope=0.840 for GRASP/Optimized and R=0.867, BIAS=-0.02, and Slope=0.662 for GRASP/Models. GRASP/Models AODF product has a slightly smaller RMSE (0.092) than GRASP/HP (0.097) and GRASP/Optimized (0.099). Even though, the GCOS requirement is initially defined for total AOD, here we also applied GCOS fraction on AODF validation based on max (±0.04, ±0.1AODF). The GCOS fraction for all AODF products is at least ~55% over land +

ocean. The GCOS fraction is highest for GRASP/Models (65.2%), which is dominant for low aerosol

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loading cases (AODF<0.2). For moderate and high aerosol loadings (AODF>=0.2), GRASP/Optimized and GRASP/HP show better performance than GRASP/Models, in terms of GCOS fraction and biases. The linear regression slope for GRASP/Models is weakest 0.662 compared to 0.892 and 0.840 for GRASP/HP and GRASP/Optimized respectively. These facts suggest a possible underestimation of fine-mode aerosol in high AOD conditions for GRASP/Models. Caution is required in the interpretation of the regression slope as these data may not meet the assumptions behind the technique; however, the results are useful in a comparative sense. The statistics for separated land and ocean are presented in Table 5. As can be seen, overall, PARASOL/GRASP AODF products show very good agreement with AERONET SDA products.

GRASP/HP AODF demonstrates best performance in terms of the highest correlation and smallest bias.

## [Figure 4]

#### [Table 5]

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The coarse-mode AOD (AODC) traditionally a difficult parameter to derive from satellite observations especially over bright land surfaces, since nadir -looking satellite measurements are not very sensitive to large particles. The validation of all archived PARASOL/GRASP AODC with AERONET SDA AODC is presented in Fig. 5. Generally, the global (land + ocean) statistical metrics for AODC are less convincing than that for AODF but still reasonable: GRASP/HP has higher correlation R (0.745) and 850 Slope (0.936) than GRASP/Optimized (R=0.689, Slope=0.748) and GRASP/Models (R=0.579, Slope=0.657). GRASP/Models retrievals show a smaller bias (0.02) and RMSE (0.109) than GRASP/Optimized (BIAS=0.04, RMSE=0.116) and GRASP/HP (BIAS=0.05, RMSE=0.123). The GCOS fraction of AODC max ( $\pm 0.04$ ,  $\pm 0.1AODC$ ) for GRASP/Models (65.4%) is higher than GRASP/Optimized (44.3%) and GRASP/HP (48.4%). In line with AODF, GRASP/Models has better performance for low 855 aerosol loading cases, which account for ~90% of the number of points. The statistics of separated land and ocean AODC validation, presented in Table 6, show a much higher correlation of retrieved AODC with AERONET over ocean. It is also interesting to note that the validation statistics for AODF seems to be superior to that for AODC over land, and the situation is reversed. This can be explained by the fact that the

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Cheng Chen 14/10/2020 12:17 Formatted: Centered Cheng Chen 17/10/2020 16:26 Deleted: 6 fine mode aerosols have higher abundance over land while coarse mode aerosol is dominant over ocean, i.e. dynamic ranges are <u>different</u>. Also, at longer wavelengths where the contribution of coarse particles to radiation is significant, the land surface is very bright while ocean surface is practically dark. Over land AODC in GRASP/HP and GRASP/Optimized products exhibit rather high BIAS of 0.05 and 0.03 correspondingly, that probably dominates the bias for the total AOD in both. For GRASP/Models product biases in AODF and AODC over land have comparable magnitudes and different signs, and therefore compensate each other in the total AOD.

## [Figure 5]

### [Table 6]

#### 870 Single Scattering Albedo

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Fig. 6 shows the validation of PARASOL/GRASP SSA (670 nm) with AERONET L2 inversion products. The SSA products in AERONET L2 database provide the values only for moderate and high AOD cases (AOD at 440 nm ≥ 0.4) to assure the highest quality of the inversion products (Dubovik et al., 2000, 2002b). Following the same strategy, PARASOL/GRASP L2 and L3 products of SSA for low AOD cases are also filtered out (Land: AOD 443 nm<0.3; Ocean: AOD 443 nm<0.02). The threshold for filtering SSA over ocean is very low because using higher values would eliminate a significant fraction of the retrievals. This low-AOD filtering is done over L2 products, and then L3 SSA is generated from filtered L2 products. The validation shows convincing correlation of all SSA PARASOL/GRASP products with those from AERONET, although due to a rather small dynamic range (mostly 0.7-1.0) of SSA, the correlation is for GRASP/HP with R=0.536 and RMSE (0.056) compared with GRASP/Optimized (R=0.511; RMSE=0.065) and GRASP/Models (R=0.324; RMSE=0.057), while GRASP/Models has a smallest BIAS (-0.02) compared to GRASP/HP (BIAS=-0.03) and GRASP/Optimized (BIAS=-0.04).</p>

Table 7 shows the statistics of PARASOL/GRASP spectral SSA (443, 670, 865, and 1020 nm) against AERONET SSA at four wavelengths (440, 675, 870, and 1020 nm). The statistics are given for Cheng Chen 26/9/2020 18:3 Deleted: difference

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combined land and ocean, because of the limited amount of validation points over ocean. The SSA correlation coefficients for GRASP/Optimized and GRASP/HP L3 products increase from 440 nm (~0.25) to 1020 nm (~0.60), which is likely due to the increased dynamic range of SSA at longer wavelengths (e.g. see Dubovik et al. (2002b), SSA at 1020 can change from very low values for biomass burning aerosol to nearly unity for desert dust). Consequently, the RMSE also increases from 440 to 1020 nm.

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In addition, Table 7 reports the statistics of SSA validation at different PARASOL AOD levels. The results clearly illustrate the improvement of retrieved SSA with the increase of aerosol abundance, in ageement with the results of AERONET sensitivity studies by Dubovik et al., (2000). For example, the correlation coefficient for GRASP/Models SSA at 670 nm with AERONET significantly improves from 0.321 for all <u>PARASOL/GRASP</u> L3 products to 0.814 for <u>PARASOL</u> AOD greater than 1.5. <u>Meanwhile</u>, the RMSE decreases from 0.056 to 0.029.

[Figure 6]

[Table 7]

#### Aerosol Absorption Optical Depth

Aerosol absorption optical depth (AAOD) is related to SSA and total AOD as:

## 905 $AAOD(\lambda) = AOD(\lambda) \times [1 - SSA(\lambda)]$

In the current PARASOL/GRASP L3 dataset, the AAOD value of each grid box (0.1° or 1° degree) is calculated based on Eq. (5) using average AOD( $\lambda$ ) and SSA( $\lambda$ ) of the grid box. Note that the PARASOL/GRASP L3 SSA( $\lambda$ ) values are aggregated based on moderate and high AOD cases (Land: AOD 443 nm $\geq$ 0.3; Ocean: AOD 443 nm $\geq$ 0.02), and again the very low threshold for filtering SSA over ocean was chosen in order to retain sufficient number of SSA and AOD retrievals. Choosing even slightly higher values would eliminate the majority of retrieval over ocean. Thus, the direct use of L3 climatology of AAOD may lead to overestimation of the global aerosol absorption, because the low AOD cases are filtered. Similarly, the AERONET L2 database provides AAOD products only for moderate and high AOD cases (AOD at 440 nm  $\geq$  0.4) to assure their highest quality (Dubovik et al., 2000).

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(5)

Cheng Chen 19/10/2020 21:47 Deleted: (>0.02) The statistics of PARASOL/GRASP spectral AAOD (443, 670, 865 and 1020 nm) validation versus AERONET AAOD (440, 675, 870, and 1020 nm) are shown in Table 8. The correlation coefficients of 920 AAOD are relatively low (0.4-0.55), which is certainly due to the low absolute value of AAOD, most cases are less than 30% of total AOD. GRASP/HP and GRASP/Models AAOD products show the RMSE equal to 0.042-0.018 from 443 nm to 1020 nm for Models, and 0.047-0.025 for HP. The BIAS is lowest for GRASP/Models AAOD: 0.00 at 440, 870 and 1020 nm and 0.01 at 670. Thus, PARASOL/GRASP AAOD provide rather useful information about global AAOD values, even the uncertainties are rather significant

925 given the generally low magnitudes of AAOD. In contrast with SSA, the attempts to analyze the AAOD accuracy for different AOD levels did not show any consistent improvement in accuracy with increase of abundance.

## [Table 8]

## 3.2 Comparison of results obtained from validation of PARASOL and MODIS aerosol products 930 against AERONET

In order to place the PARASOL/GRASP validation results into perspective, here we compare PARASOL/GRASP ability to retrieve AOD, AE, AODF and AODC with other satellites. Specifically, these products from MODIS, PARASOL/Operational and PARASOL/GRASP products are validated using the same approach for the entire 2008 year and validation results were compared. MODIS aerosol products have been extensively evaluated globally by the MODIS team in multiple studies (Gupta et al., 2018; Levy et al., 2010, 2013, 2018; Lyapustin et al., 2018; Sayer et al., 2013, 2014, 2019), and PARASOL/Operational aerosol products have been evaluated in Bréon et al. (2011); the present analyses is performed for reader convenience and consistency of methodology across products. We confirmed that the statistic metrics that

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studies. This section is therefore focusing on a comprehensive evaluation of the consistencies and differences between PARASOL and MODIS aerosol products using examples from one year. The year 2008 was chosen because it presents a generally good statistics of observations and all types of aerosol are

we found for MODIS and PARASOL/Operational aerosol products validation in 2008 is similar to these

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Unknown Field Code Changed Utilisateur Microsoft O..., 2/11/2020 12:59 Formatted: English (UK) Utilisateur Microsoft O..., 2/11/2020 12:59 Formatted: English (UK) 945 clearly present. The validation figures for the satellite products over land and ocean are presented separately, because over land there are three MODIS products (DT, DB, and MAIAC), while only DT product is provided over ocean. MAIAC products cover some land-containing ocean tiles, <u>however due to limited coverage of these retrievals</u>, we do not consider MAIAC ocean products here. PARASOL/Operational AODF products are provided over land and ocean, and total AOD products only
950 over ocean.

Fig. 7 shows validation results for AOD at 550 nm for 3 PARASOL/GRASP products over land with collocated AERONET measurements. Fig. 8 shows the validation results for MODIS DT, DB and MAIAC AOD products over land. The products called MAIAC 0.1 and MAIAC 0.01 correspond to the MODIS MAIAC original product aggregated to 0.1° and 0.01° grid boxes, respectively. In general MODIS 955 products have more matched points than PARASOL products due to MODIS' wider swath and higher spatial resolution of measurements allowing better cloud detection. From low to high values the sequence obtained global correlation coefficients is: 0.870 (DB), 0.874 (MAIAC 0.01), 0.875 of (GRASP/Optimized), 0.895 (MAIAC 0.1), 0.898 (DT), 0.908 (GRASP/HP) and 0.924 (GRASP/Models). The GCOS fraction sequence is 28.8% (GRASP/Optimized), 32.4% (GRASP/HP), 46.1% (DT), 48.1% (MAIAC 0.01), 48.8% (DB), 52.8% (MAIAC 0.1), and 53.2% (GRASP/Models). The high to low RMSE 960 sequence is: 0.157 (GRASP/HP), 0.150 (GRASP/Optimized), 0.126 (DB), 0.121 (GRASP/Models), 0.120 (DT), and 0.112 (MAIAC 0.1). The large to small total BIAS sequence is: 0.06 (GRASP/HP), 0.04 (GRASP/Optimized), -0.03 (MAIAC 0.01 and MAIAC 0.1), 0.02 (DT), -0.01 (DB) and 0.00 (GRASP/Models). The low to high sequence of regression slope values is: 0.780 (GRASP/Optimized), 965 0.793 (MAIAC 0.1), 0.794 (MAIAC 0.1), 0.841 (DB), 0.938 (GRASP/HP), 0.988 (DT), and 0.989 (GRASP/Models). The results illustrate that the overall accuracy of these AOD products are generally comparable on a global scale. Note, however, that different products may have different regional strengths and weaknesses (e.g. Sayer et al., 2014), motivating the mapped analysis later. The GRASP/Models AOD yields overall the largest number of the best statistical indicators over land: with the highest correlation Cheng Chen 30/9/2020 12:17 Deleted: though as these are spatially complete

(R=0.924), the highest GCOS fraction (53.2%), correlation slope (0.989) and the smallest total bias (BIAS=0.00). The detailed statistics of PARASOL and MODIS AOD products against referenced collocated AERONET AOD at 550 nm over land and ocean are presented in Table 9.

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# [Figure 7]

## [Figure 8]

PARASOL/GRASP Fig. 9 presents validation of (Optimized, HP and Models), PARASOL/Operational and MODIS/DT AOD products versus collocated AERONET measurements over ocean in 2008. The detailed statistic metrics are presented in Table 9. The matching methodology is the same as described in section 2.4. The total matched points in 2008 are ranging from minimum 980 GRASP/Optimized 116 to maximum MODIS/DT 218. In general, all AOD products show high correlation over ocean, of which GRASP/Models has the highest R (0.963), following by Operational (0.954), DT (0.952), GRASP/Optimized (0.950) and GRASP/HP (0.947). The high to low sequence of RMSE is 0.092 (GRASP/HP), 0.089 (GRASP/Optimized), 0.081 (DT), 0.077 (Operational) and 0.061 (GRASP/Models). The slopes are quite similar: 1.165 (Operational), 1.145 (GRASP/Optimized), 1.074 (GRASP/HP), 0.965 985 (GRASP/Models) and 0.974 (DT). Overall, GRASP/Models show slightly better bias (0.02) and GCOS fraction (62.9%), following by DT (BIAS=0.03, GCOS fraction=55.0%), Operational (BIAS=0.03, GCOS fraction=52.2%), GRASP/Optimized (BIAS=0.06, GCOS fraction=42.2%) and GRASP/HP (BIAS=0.07, GCOS fraction=26.6%). Altogether, GRASP/Models, PARASOL/Operational and DT AOD yield quite similar performance over ocean with a better statistics than GRASP/Optimized and GRASP/HP AODs that

990 correlate well with AERONET measurements but present a significant positive bias (0.06-0.07).

## [Figure 9]

#### [Table 9]

In order to obtain more information about the quality of the retrieval products over different land surfaces, the statistics of satellite validation against AERONET were also analyzed separately for different land covers. Table 10 shows the statistic metrics for land surfaces with different Normalized Difference Vegetation Index (NDVI). The statistics are presented for several categories: bare soil/desert surfaces Deleted: correlation

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Cheng Chen 14/10/2020 12:21 Formatted: Centered Cheng Chen 17/10/2020 16:30 Deleted: 10 of vegetation (0.4≤NDVI<0.6 and NDVI≥0.6). The global NDVI dataset is adopted from GRASP/Models</li>
L3 annual mean products for year 2008 (Fig. 10). The <u>statistic metrics in Table 10 show that, in general, all</u> products show better performance over surface type with 0.2≤NDVI<0.6 than bright, bare surfaces (NDVI<0.2), and somewhat better than for dense vegetation surface (NDVI≥0.6). Overall the AOD product of GRASP/Models seems to show the best correlation with AERONET, with highest R over 3 of 4 surface</li>
classes. Over bright surfaces (NDVI<0.2), GRASP/HP has a highest R (0.915), but also rather high BIAS of 0.06. The GRASP/Models AOD also has zero BIAS for 3 surface classes except the dense vegetation surface (NDVI≥0.6), where GRASP/Models AOD has total BIAS of 0.03, higher than that in any MODIS AODs.</li>

(NDVI<0.2); mixture of bare soil and vegetated surfaces (0.2≤NDVI<0.4); surfaces covered different types

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## [Figure 10]

#### [Table 10]

Fig. 11 shows the validation of AE for PARASOL/GRASP and MODIS DT and DB products over land versus collocated AERONET measurements. The MODIS AE for DT and DB products were calculated based on 470 and 660 nm that are reported in both products; an equivalent for AERONET was calculated using AOD interpolated to 470 and 660 nm. PARASOL/GRASP products contain AOD at 440 1015 and 870 nm, therefore AE (440/870) was directly used for validation PARASOL/GRASP results. MAIAC AE was not included because MAIAC reports AOD at two rather close wavelengths 470 and 550 nm and calculation of AE using these such close channels could produce substantial uncertainties in AE. The threshold of satellite AOD (550 nm) > 0.2 was used in validation of AE over land and ocean. In general,

PARASOL/GRASP (Optimized, HP and Models) AE <u>agree notably better with AERONET than MODIS</u> (DT and DB), which is likely caused by the lower information content in regards to aerosol size in mono-

1020 (DT and DB), which is likely caused by the lower information content in regards to aerosol size in monoviewing MODIS observations. In addition, both DT and DB algorithms rely on climatology for the aerosol model selection, i.e. AE is rather predetermined than retrieved to some extent. For example, although AOD over land is reported by DT at 470 nm and 660 nm, the spectral dependence of the DT land retrieval is Cheng Chen 20/10/2020 14: **Deleted:** correlation

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mostly imposed by assumed aerosol models. The DT team makes a specific point of not reporting AE over land for that reason, and at best the spectral dependence might allow a binary inference of either fine mode or coarse mode dominated particles, but not a quantitative measure of the true spectral dependence. The DT over ocean algorithm has greater flexibility in its mixing of models and does return a quantitative AE. The weaker performance of GRASP/Models approach compare to GRASP/HP and GRASP/Optimized is caused by the limitation of maximum and minimum AE values allowed by the mixture of aerosol components used, even though GRASP/Models approach allows mixing of different components freely with no location specific constraints. As a result, GRASP/Models tends to overestimate AE for large

GRASP/Models AE products are less appealing than those from GRASP/Optimized and GRASP/HP in 1035 terms of evaluation metrics. GRASP/HP tends to provide the most reliable AE products over land.

# [Figure 11] [Figure 12] [Table 11]

particles (low AE values), and underestimate AE for small particles (high AE values). Hence,

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Fig. 12 presents the validation of AE over ocean from PARASOL/GRASP, PARASOL/Operational and MODIS/DT products against AERONET measurements. PARASOL/Operational AE was calculated based on 670 and 870 nm that are reported in the product, and AE (670/870) of AERONET was calculated using AOD interpolated to 670 and 870 nm. Although there are not many available points, the satellite derived AE over ocean are much better than that over land. GRASP/Models show R (0.949) higher than Operational (0.891), GRASP/HP (0.890), GRASP/Optimized (0.840) and DT (0.832). GRASP/Models and 1045 MODIS DT AE show an overestimation for large particles. Operational AE tends to overestimate both for large and small particles. At the same time, GRASP/HP AE correlation has the slope closer to 1:1 line with

AERONET AE than other products, with the best linear fitting slope (0.810) and intercept (0.051). The statistic metrics of AE validation over land and ocean are listed in Table 11.

AODF is often used to estimate anthropogenic aerosol climate effects (Bellouin et al., 2005) and 1050 surface air quality (e.g. PM<sub>2.5</sub>) (Zhang and Li, 2015). MODIS started to report fine mode weighting 33

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parameter (η) in the products from the second generation DT operational algorithm (Levy et al., 2007b), though η is weighted for reflectance not for AOD. Consequently η over land is a diagnostic that has little
physical meaning and the resulting AODF and AODC do not have physical meaning and generally are not recommended to be used. Therefore, it is not considered in the analysis. However, over ocean, based on single scattering approximation, η is also weighted for AOD (Remer et al., 2005). Therefore, MODIS fine and coarse mode AOD at 550 nm over ocean are derived according to the equations below:

$$AODF = AOD \times \eta \tag{6}$$

$$AODC = AOD \times (1.0 - \eta)$$
<sup>(7)</sup>

Fig. 13 shows the validation of AODF at 550 nm for PARASOL/GRASP and 1060 PARASOL/Operational AODF products over land against AERONET AODF products from the SDA algorithm. It's noticeable that the AODF products over land are only available from PARASOL MAP measurements. The results in Fig. 13 indicate the PARASOL/GRASP and PARASOL/Operational AODF products are in good agreement with AERONET SDA products, for example, R>0.86. The GCOS fraction of AODF for PARASOL/GRASP and PARASOL/Operational products are at least 50%. GRASP/HP 1065 AODF shows the best correlation among all four AODF products over land, with rather similar performance for GRASP/Optimized AODF.

The AODF validation over ocean is shown in Fig. 14, and statistical metrics over land and ocean are presented in Table 12. GRASP/Optimized and GRASP/HP AODF show generally consistent performance over ocean and over land, with correlation R around 0.9, while the BIAS for GRASP/Optimized and GRASP/HP AODF is higher over ocean than over land. At the same time, GRASP/Models AODF shows significant improvement over ocean, for example, the fitting line is much closer to 1:1 (dotted line), and the RMSE decreased dramatically. PARASOL/Operational AODF shows a slight decrease of R from land (R=0.886) to ocean (R=0.780), also reported in Bréon et al. (2011), while the fitting line, RMSE and BIAS show improvement from land to ocean. This is likely due to higher information content about aerosols in satellite observations over dark ocean surfaces compared to brighter land surfaces.

## [Figure 13] [Figure 14]

## [Table 12]

The validation of AODC over land and ocean are shown in Figs. 15 and 16, respectively. Even 1080 though AODC products over land are only provided for PARASOL/GRASP, for completeness we present this over land for year 2008 in Fig. 15. Similarly to the results from the total PARASOL/GRASP archive, AODC over ocean is more accurate than over land. The overall best results of AODC are provided by GRASP/HP with highest R (0.771) and the best linear fitting (slope is reaching 1 and intercept is close to 0) over land. Yet, the BIAS of GRASP/HP AODC is 0.05, which is higher than GRASP/Models (0.01) and 1085 GRASP/Optimized (0.03), which result in higher GCOS fraction for GRASP/Models AODC (63.7%) than Deleted: s GRASP/HP (45.8%) and GRASP/Optimized (45.6%). At the same time, as mentioned above, over dark ocean the sensitivity of the observed signal to aerosol is stronger allowing for retrieval of particle size information that is more challenging over land. The GRASP/Models AODC shows the best R (0.966) and RMSE (0.040) while MODIS/DT AODC has the smallest bias (0.00) against AERONET over ocean, 1090 following by Operational (0.01), GRASP/Models (-0.01), GRASP/Optimized (0.03), and GRASP/HP (0.05). Although AODC is not included in PARASOL/Operational product list, over ocean we subtract AODF from total AOD to obtain Operational AODC, which shows a rather good agreement with AERONET (R=0.936, Slope=0.971, RMSE=0.045, BIAS=0.01). However, over land, only AODF is provided in the PARASOL/Operational product.

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[Figure 15] [Figure 16] [Table 13]

The statistics in comparison of each single product vary due to the differences in product coverage: coverage of MODIS/AQUA is wider than PARASOL; at the same time, the products have different limitations and availability: Cheng Chen 14/10/2020 12:: Formatted: Centered

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- MODIS/DT has limited products over bright land surfaces, and AODF and AODC are only available over ocean;
- MODIS/DB and MAIAC AOD products are only over land, and do not include AODF and AODC;
  - PARASOL/Operational over land provides only AODF;
  - Quality screening is different (even between PARASOL/GRASP products).

Therefore, the approaches chosen in this paper for considering all above factors could have some effects on the results and their interpretation. At the same time, the correlations with AERONET obtained in these studies for known products including MODIS DT, DB and MAIAC and PARASOL/Operational in general agree with the results of previously mentioned studies.

## 3.3 Evaluation of PARASOL and MODIS validation results over different AERONET sites

- In this section, we compare the validation metrics of PARASOL/GRASP and MODIS aerosol products over spatially distributed AERONET sites. PARASOL/Operational AOD products are provided over ocean only, hence are not included in this section. The AOD validation was conducted over all AERONET sites that had available data in 2008. At the same time, and to increase statistical robustness only sites with at least 10 matchup points were included in the analysis. However, the different products can also have different number of matchup points over different AERONET sites due to various factors (as
- 1120 discussed previously). Therefore, to evaluate the validation performance of different products, the percentage (%) of the cases when the product of each algorithm showed the best statistic metrics, observed among all the products (e.g. the highest R, GCOS Fraction, and the lowest RMSE, BIAS, etc.) was used as an indicator for the performance evaluation.

Fig. 17 shows the percentage score for each algorithm at AERONET sites for statistical metrics R,
 RMSE, BIAS and GCOS Fraction respectively. The detailed statistics for the performance of each AOD products is shown in Fig. 18 (only the 1<sup>st</sup> ranking statistics over each site are present in the maps.). All PARASOL/GRASP products have fewer sites with at least 10 matchup points than MODIS AOD products.

There are 102, 124, and 95 sites having sufficient matchup points for GRASP/Optimized, GRASP/HP and GRASP/Models respectively, lower than DT (153), DB (172), MAIAC 0.1 (169) and MAIAC 0.01 (172)

- 1130 by 20%~45%. Regarding the correlation coefficient R, GRASP/Models, DT and MAIAC\_0.1 are the 3 algorithms showing higher scores for 37.9%, 28.1% and 24.9% sites where respective products were provided. As shown in Fig. 18a, these 3 algorithms show good performance worldwide, e.g. North America, Europe and East Asia. There are no MODIS AOD products showing the best R over Australia (only 4 sites are available there). The 3 GRASP algorithms show high percentage for products over dust and biomass 1135 burning regions, e.g. South America, southern Africa, central Africa, central Australia, etc. At the same
- time, the GRASP/Optimized and GRASP/HP AOD products performed less well over North America.

# [Figure 17]

### [Figure 18]

In terms of the percentage of sites with the best RMSE, GRASP/Models and MAIAC\_0.1 are the top 2 algorithms showing the best RMSE results over 60.0% and 33.1% of AERONET sites with available GRASP/Models and MAIAC\_0.1 retrievals. Thus, overall, all these results indicate that GRASP/Models presents a strong ability to provide AOD that agrees well with AERONET measurements. Both GRASP/Models and MAIAC\_0.1 show the best RMSE over Europe and North America (Fig. 18b), which also have the highest density of AERONET sites. GRASP/Models shows the best BIAS over 36.8% sites, followed by DB (27.3%) and MAIAC\_0.01 (23.3%). For the best GCOS Fraction, GRASP/Models leads this for 57.9% over its total 95 sites. After, MAIAC\_0.1 has the highest GCOS Fraction for 30.8% over total 169 sites. In Fig. 18d, the best GRASP/Models sites are globally distributed. Over the Eastern United States, DB and MAIAC\_0.1 products tend to have more sites with the best GCOS Fraction.

Using a similar concept as the AOD analysis above, the PARASOL/GRASP and MODIS (DT and DB) AE validation over AERONET sites were compared. Only cases with satellite AOD (550 nm) > 0.2 were included in the analysis; due to the reduced data volume from this threshold, the requirement on minimum matchups was reduced to 5. Fig. 19 shows the detailed statistics for the performance of each AE products. Fig. 20 shows the best performing algorithm at each site according to R and RMSE respectively.

In general, GRASP/HP and GRASP/Optimized AE products outperform the other AE products in the site 1155 level validation. The best sites are globally distributed (see Fig. 20). There are 44.1%, 38.6% and 34.7% sites showing the best R for GRASP/Optimized, GRASP/HP and GRASP/Models, somewhat higher than DT (12.0%) and DB (8.2%). GRASP/HP AE has the best RMSE over 43.0% AERONET sites, higher than GRASP/Optimized (34.3%), DB (28.6%), GRASP/Models (24.5%) and DT (17.0%).

## [Figure 19] [Figure 20]

#### 4 Inter-comparison of satellite products at global scale

This section presents the inter-comparison of different satellite products for the year 2008 data on a global scale, i.e. not only over AERONET sites. Specifically, we want to know if the consistency of the satellite products remains the same in the areas where no AERONET observations are available. In the first part, we compare PARASOL/GRASP and PARASOL/Operational at a spatial resolution of 0.2° x 0.2°, which represents a compromise between PARASOL/Operational (18.5 km) and PARASOL/GRASP Level 3 (0.1°) resolutions. In the second part of this section, the global inter-comparison is done between PARASOL/GRASP and MODIS aerosol products in a spatial resolution of 0.1° x 0.1°, close to the DT and DB product native resolution of 10 km, and use MAIAC\_0.1 data that is of similar resolution. Only

1170 GRASP/HP and GRASP/Models products for PARASOL/GRASP are used in the consideration of this section since GRASP/Optimized shows rather similar results to GRASP/HP. Since the focus of this section is global pixel-to-pixel comparison of satellite aerosol products, we use all available data of the highest quality for each dataset (Table 2).

## 4.1 Comparisons between PARASOL/GRASP and PARASOL/Operational aerosol products

To begin, we investigate two independent aerosol products derived from PARASOL measurements, PARASOL/GRASP and PARASOL/Operational, globally for 2008. As mentioned above, PARASOL/Operational provides only AODF over land, while over ocean AOD, AE and AODF are available. We subtract AODF from total AOD to obtain Operational AODC over ocean.

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Table 14 presents the pixel-to-pixel statistic metrics (R, Slope, Intercept, RMSE and BIAS) between Operational and GRASP aerosol products (note here the BIAS should be interpreted as an offset rather than true bias as the "truth" is unknown; we retain the name of the metric for consistency with the earlier analysis). We took Operational products as a reference as these were the original PARASOL aerosol products released by AERIS/ICARE; hence, the BIAS is defined as GRASP - Operational. All the statistics

- 1185 for AOD, AODF and AODC are given for the midvisible wavelength (550 nm), while AE is calculated based on 670 and 870 nm. The statistical metrics are reported both for global comparisons and over AERONET pixels only (the numbers in the brackets). It can be seen from Table 14 the global comparison between PARASOL/GRASP and PARASOL/Operational is rather consistent for AOD over ocean and AODF over land, for which, the global pixel-to-pixel correlations between GRASP/HP, GRASP/Models
- and Operational products are generally higher than 0.85 based on more than 5 million pairs. However, the agreement of AODF over ocean decreases to 0.63-0.73 for R. The slight decreasing of correlation against AERONET from land to ocean for Operational AODF products is also recorded in Table 12 and previous study by Bréon et al. (2011). The AODC over ocean for the Operational product is derived from AOD and AODF, hence, the number of matched pairs is lower than for AODF. The overall agreement has a
- 1195 correlation coefficient of ~0.7. GRASP/HP AODC is ~0.05 higher than Operational, but the difference between GRASP/Models and Operational is ~0.0, which are in line with the validation against AERONET in Table 13. The pixel-to-pixel agreement for PARASOL/GRASP and PARASOL/Operational AE is less convincing (R<0.6) than any other parameters, even though they are all well correlated with AERONET (R>0.8) over ocean. One possible reason is that the AE here is calculated at different wavelengths (670 and
- 870 nm) than for the comparisons with AERONET (470/660 nm and 440/870 nm). Besides, the <u>decrease of</u>
   AE agreement for global correlation (R) compared to that over AERONET pixels is more notable than
   other parameters. This may explain that the AE products resulting from LUT-based algorithms are more
   determined by climatological assumptions about the aerosol models than retrieved.

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#### [Table 14]

## 4.2 AOD comparisons between PARASOL/GRASP and MODIS products

In order to further clarify the level of consistency of satellite products (PARASOL/GRASP and MODIS), the global correlations of different satellite products were extensively analyzed for the year 2008 at a spatial resolution of 0.1° x 0.1°. Fig. 21 shows the seasonal pattern of AOD (550 nm) from PARASOL (GRASP/HP and GRASP/Models) and MODIS (DT, DB, and MAIAC) products. Any grid box with less

- than 3 measurements for a season was omitted. Fig. 22 shows the differences of <u>mean\_AOD</u> (550 nm) by season between PARASOL and MODIS aerosol products using GRASP/Models as the reference. A positive value indicates that the MODIS product had a higher mean value. Since the analysis in Section 3 suggested that the AOD products over land and ocean from the GRASP/Models processing have the lowest
- biases, this was used as a reference product in Fig. 22. Since GRASP/HP AOD shows non-negligible BIAS
   (land: +0.06; ocean: +0.07) from AERONET validation (see Table 9), in order to show the intrinsic, the
   bias was subtracted from the GRASP/HP AOD products before obtaining the seasonal differences shown in
   Fig. 22.
- In addition, the global correlations between different satellite products and GRASP/Models data at 550 nm were calculated for the complete year 2008. Also, in order to evaluate the consistency of different MODIS products over land, the inter-comparisons were done against MAIAC AOD (Land) product chosen as a reference, as MAIAC provides the most universal coverage over land. Table 15 presents the pixel-topixel statistic metrics (R, Slope, Intercept, RMSE and BIAS) between AOD products compared to the reference of GRASP/Models (Land and Ocean) and MAIAC AOD (Land) products. The statistical metrics are reported both for global comparisons and over AERONET pixels only (numbers in brackets).

## [Figure 21]

## [Figure 22]

## [Table 1<u>5</u>]

Each of these global correlations was based on several dozens of millions of pairs, and less noisy compared to the AERONET correlations (based on only a few thousand points). In spite of this significant

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Cheng Chen 14/10/2020 12:23 Formatted: Centered difference in volume, the outcome of the global satellite comparisons is rather consistent with the results of

- 1245 validation against AERONET. For example, all AOD products are in close agreement over ocean, with the correlation coefficients above 0.9 and slope lines close to 1:1 (Table 15). Specifically, the three aerosol products (GRASP/HP, GRASP/Models and DT) over ocean agree with R>0.92 for any two products. Also, in line with the validation over AERONET sites, GRASP/HP AOD (550 nm) consistently has a positive offset ~0.05-0.16 from low to high AOD conditions with respect to GRASP/Models. DT and 1250 GRASP/Models AOD show good agreement over ocean, R=0.92 for all points and R=0.97 for AERONET
- pixels, in addition, the BIAS (DT–GRASP/Models) equals to -0.01 for all points and 0.00 for low AOD (<0.2), while the negative BIAS of -0.06 appears when AOD is greater than 0.7. Statistics over ocean rely on ~65 million pairs between GRASP/HP and GRASP/Models, and ~32 million pairs between DT and GRASP/Models.
- However, over land surfaces the situation is quite different, and MODIS/MAIAC and DB AOD products show evidently better agreement with GRASP/Models over AERONET pixels than the rest of globe. The correlations over AERONET pixels both for MAIAC versus GRASP/Models and DB versus GRASP/Models are of ~0.89 that is generally in line with the correlation coefficient values with AERONET shown in Table 9. In a contrast, the corresponding correlation coefficients decrease to 0.76 and 0.77 for global statistics. The other statistical parameters (e.g. Slope, Offset, RMSE and BIAS) showed the
- same trend, indicating a better agreement over AERONET pixels. For comparisons of GRASP/HP and DT versus GRASP/Models AOD such tendency is not evident. Even though, the correlation coefficient drops from 0.90 over AERONET to 0.85 globally, the rest of statistical indicators do not show significant changes, whether over an AERONET site or elsewhere. It is interesting to note that MODIS products show better agreement (especially in correlations) with other MODIS products over AERONET stations and globally than between PARASOL and MODIS products over AERONET stations and globally (Table 15).
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This phenomenon can be explained by several factors. First, the inputs from the two satellites differ

significantly. The multi-angle polarization information from PARASOL offers algorithms many more

degrees of freedom from which to constrain environmental factors and invert aerosol parameters than does

- 1270 a single view radiometer like MODIS. Second, because of this extra information the PARASOL/GRASP retrievals do not have location specific assumption about aerosol and conduct their retrievals in the exactly the same manner globally. In contrast, all three MODIS retrievals use some regional assumptions over land about aerosol types, surface properties, etc. Even though each algorithm's assumptions are different, the need for a priori constraints could draw the MODIS products closer together. Therefore, the similarities in global performance of three algorithms can probably be explained by somewhat similar a priori assumptions about aerosol types, etc. used in MODIS algorithms. Third, as can be seen from the Table 15 and Fig. 21, GRASP/Models, GRASP/HP and MAIAC have wider coverage over land than DB and DT, because of the lack of retrievals over bright surfaces for DT and reduced number of retrievals over dark vegetation for DB (although some of this was improved in DB Collection 6.1; Sayer et al., 2019).
- Specifically, for the year 2008, there are more than 64 millions of pairs MAIAC/GRASP/Models AOD over land, which is much higher than the number of pairs obtained with other two AOD products. Thus, the collocation statistics for MAIAC/GRASP, DT/GRASP, DB/GRASP as well as MAIAC/DT and MAIAC/DB were based on different data sets. Fourth, the different representation of various natural conditions in the global statistics and statistics over AERONET can be non-identical and, therefore, the average performance indications can differ. For example, there is only a certain fraction of AERONET sites in desert areas while land cover with bright surface may have notably higher or lower fraction in global statistics. Correspondingly, if the product agreement is non-identical over different land surfaces, then the

statistics with different representations of various surfaces can differ.

In order to explore the last factor, the statistics of the comparisons were sorted by land surface type. 1290 The Tables 16 and 17 show pixel-to-pixel statistic metrics with reference AOD from GRASP/Models and MAIAC respectively, over different land coverage using four classes of land surface by NDVI (as before, NDVI<0.2, 0.2≤NDVI<0.4, 0.4≤NDVI<0.6, and NDVI≥0.6).

[Table 16]

## [Table 1<u>7</u>]

1295 Tables 16 and 17 show that over very bright land surfaces (NDVI < 0.2), the global correlations between MODIS (especially DB and MAIAC) with PARASOL/Models products were significantly lower than over other surfaces and showed a most notable drop (>0.1) in global correlations compared to the correlation over AERONET sites. Such a large drop was not seen between different PARASOL products or between different MODIS products. Therefore, these differences are likely related to the fact that MODIS retrievals rely on regional climatological aerosol assumptions or surface assumptions derived from 1300 atmospheric correction at (unevenly-distributed) AERONET sites while in PARASOL/GRASP retrievals no location specific assumptions are used. Another issue maybe related is that MODIS has much higher spatial resolution for cloud detection than PARASOL. The possible sub-pixel cloud contamination for PARASOL may affect the global inter-comparison statistics, since the validation against AERONET brings 1305 additional cloud clearing filter from AERONET. As a result, PARASOL/GRASP retrievals are expected to be rather consistent globally, while MODIS retrievals are more closely tied to AERONET statistics and may perform less well in the areas with a lack of AERONET sites. At the same time, the fraction of pairs over bright surfaces in inter-satellite product comparisons is higher than in AERONET statistics since there are only a limited number of AERONET sites in desert areas. This latter statement does not necessarily apply to MODIS DT because it often does not retrieve over deserts; however, although the sample size is 1310 very small, Table 16 shows that it actually matches GRASP/Models less well at AERONET sites than

globally for NDVI < 0.2.

Interestingly, the maps in Fig. 22 of seasonal AOD difference indicate lower AOD (550 nm) for PARASOL/Models over bright surfaces compared to MODIS products, while the global comparisons of 1315 PARASOL/Models and MODIS DB and MAIAC products did not show significant BIAS in AOD (550 nm). At the same time, the global comparisons (Table 16) between PARASOL/Models and MODIS DB, MAIAC show a significant BIAS for different ranges of AODs. MODIS DB and MAIAC had a positive BIAS of ~0.06-0.04 for the situation with lower aerosol loadings (AOD 550 nm<0.2) and a notable

negative BIAS (0.02~0.06) for moderate (0.2 < AOD 550 nm < 0.7) and especially large for high aerosol

- 1320 loadings (AOD 550 nm > 0.7) that reached ~0.3. A very similar tendency can be seen from the statistics of validation against AERONET in Table 10: both PARASOL/Models and MODIS/DB have very small BIAS of -0.01, while the distribution of BIAS is quite different for the situations with different loadings: 0.01 and 0.03 for low AODs, -0.03 and -0.05 for moderate AODs and for high AOD 0.01 and -0.16. This suggests that the observed positive differences when MODIS/DB and MAIAC show higher AOD over bright
- surfaces occurs mainly during low AOD conditions. This conclusion is supported by the fact that seasonal means from all products do not show high AOD over the northern Sahara between 20°N and 30°N latitude. Also, both DB and MAIAIC show significant underestimation of AOD over the Taklamakan desert where seasonal mean AOD retrieved by PARASOL is high, which agrees with the negative offset between MODIS DB, MAIAC and PARASOL/Models products over bright and bare soil land surfaces (NDVI<0.4).</p>
- The negative BIAS between MODIS and PARASOL products is clearly seen on the maps of 1330 seasonal AOD from different products for African biomass burning events. The results of correlation analysis over green vegetation (NDVI  $\geq$  0.6) in Table 16 also show a significant negative BIAS in all MODIS products compared to PARASOL/Models over green vegetation that increases for medium and high aerosol loading. The validation against AERONET in Table 10 shows the highest BIAS of 0.06 to 0.07 for PARASOL/Models is over green vegetation (NDVI  $\geq$  0.6) when 0.2 < AOD < 0.7, while the 1335 MODIS products tend to be less biased (DT BIAS = 0.03) or negatively biased (MAIAC BIAS = -0.04 to -0.06, and DB BIAS = -0.04) for this surface type and AOD range. This pattern continues for DB and MAIAC through all the vegetated surfaces with NDVI > 0.2. MODIS DB and MAIAC continue to be more negatively biased against AERONET for moderate to high aerosol loading than PARASOL/Models is. Thus, the results suggest that observed differences for African biomass burning events can be explained by 1340 two potential reasons: a combination of overestimations of AOD by PARASOL/GRASP retrievals and underestimation of AOD by MODIS products for cases of moderate to high aerosol loading. However, the DT retrievals also show this negative bias against PARASOL/GRASP in the African biomass burning

(Figure 22), but do not follow the same trends against MODIS as DB and MAIAC. Other factors, such as

1345 differences in cloud-screening, data amount, aggregation and quality screening approaches must also contribute to these differences and need to be investigated in future analysis.

### 4.3 AE comparisons between PARASOL/GRASP and MODIS products

The seasonal pattern of AE from PARASOL (GRASP/HP and GRASP/Models) and MODIS (DT and DB) products is presented in Fig. 23, as well as, AE differences by season between PARASOL and MODIS aerosol products in Fig. 24. Table 18 shows the global pixel-to-pixel statistic metrics between AE products based on references of GRASP/HP; in the brackets, the values corresponding to validation results over AERONET pixels only. As before, the statistic metrics split into four classes of land surface by NDVI are presented in Table 19. The GRASP/HP AE products are chosen to be a reference taking into account the highest obtained correlation in the validation with AERONET in the Section 3. Again, note that 1355 although AOD over land is reported by DT at 470 nm and 660 nm, the spectral dependence of the DT land retrieval is mostly imposed by assumed aerosol models, and thus DT AE over land is at most a binary indication of fine and coarse particles, and not a quantitative parameter. We expect no correlation with

GRASP/HP over land. AE over land from DB is similarly prescribed, not retrieved, when AOD < 0.2 (Hsu et al., 2013). On the other hand, the DT AE over ocean is a true quantitative measure.

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[Figure 23] [Figure 24] [Table 1<u>8]</u> [Table <u>19</u>]

The differences between PARASOL/GRASP and MODIS DT and DB AE products are pronounced

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in all comparisons. From Fig. 23, the seasonal variations for DB and DT are minor, which likely implies utilization of similar climatological information in the DB and DT algorithms. Even though the differences for GRASP/Models and GRASP/HP shown in Fig. 24 are not small (mainly due to the limited dynamic range of aerosol components used in the GRASP/Models approach), the overall pixel-to-pixel correlation

between GRASP/Models and GRASP/HP is the highest between any two products (0.70 over land, 0.74

- over ocean), and the RMSEs are 0.39 over land and 0.45 over ocean. The correlations for AE over land between MODIS DT and DB AE versus GRASP/HP are lower than 0.5 and RMSEs are higher than 0.59 for all land surface types (Table 19), which is not surprising for the aforementioned reasons. Over ocean, all available products (GRASP/HP, GRASP/Models and DT) show good agreement with AERONET measurements, with R>0.8 (Fig. 11 and Table 11), however, the pixel-to-pixel correlation between DT and
- 1375 GRASP/HP for ocean pixels globally decreases to 0.46, with RMSE=0.53. The cause of the drop for global statistics is presently unknown. It could be due to assumptions in the DT retrieval, but could also be linked to differences in calibration between POLDER and MODIS, as AE is particularly sensitive to nuanced spectral changes in calibration in the lower-AOD conditions often seen over ocean.

## 4.4 AODF and AODC comparisons between PARASOL/GRASP and MODIS products

This section compares AODF and AODC at 550 nm from PARASOL/GRASP (GRASP/HP and GRASP/Models) and MODIS DT algorithms. As discussed earlier, the quantitative fine mode fraction (η) provided by the DT algorithm can be used to derive AODF and AODC only over ocean. Therefore, the comparison of AODF and AODC over land is between GRASP/HP and GRASP/Models. The seasonal distribution of AODF and AODC are shown in Fig. 25 and Fig. 27 respectively. The seasonal differences between GRASP/Models, DT and GRASP/HP are shown in Fig. 26 (AODF) and Fig. 28 (AODC). GRASP/Models AODF is higher than GRASP/HP over dust source and downwind regions, while it is lower than GRASP/HP over biomass burning and urban areas, which is consistent with the validation versus AERONET measurements in Figs. 13-16.

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[Figure 25] [Figure 26] [Figure 27] [Figure 28]

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Globally, GRASP/Models and GRASP/HP AODF show a consistent agreement over land (R=0.87, BIAS=-0.01) and ocean (R=0.89, BIAS=0.00), as presented in Table 20. MODIS/DT AODF and AODC 1395 over ocean have good agreement with GRASP/HP with R 0.86 and 0.84 respectively. GRASP/Models AODC shows a better agreement with GRASP/HP over ocean than over land, while differences are less pronounced, R of 0.89 and 0.71, respectively. As was mentioned above, this tendency can be explained by a stronger sensitivity of the observed signal to aerosol over dark ocean surface. Another interesting tendency is that correlations for AODF over land are generally higher than for AODC, while over ocean the 1400 situation is inverse and the correlations are higher for AODC, especially over AERONET. This can probably be explained by the two facts that dominating oceanic aerosol has a pronounced coarse mode and that at the longer wavelengths, where the contribution of coarse mode is the strongest, the ocean is practically dark. The land reflectance is, however, higher than ocean at long wavelengths, even for 1405 relatively dark vegetated surfaces. The statistics of pixel-to-pixel comparison (GRASP/HP and GRASP/Models) over different land surface types, as discriminated by different NDVI categories, are also reported in Table 22 (AODF) and Table 23 (AODC).

> [Table 2<u>0]</u> [Table 21] [Table 2<u>2</u>] [Table 23]

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In conclusion, the differences in more detailed aerosol characteristics including AE, AODF and AODC (Tables 18-23) derived from PARASOL and MODIS are pronounced over both land and ocean. This is in contrast to the results for the total AOD from PARASOL and MODIS, which are close over ocean and in a reasonable agreement over land. This conclusion can likely be generalised by the fact that retrieval accuracy of detailed aerosol properties is expected to be significantly higher from MAP products than from mono-viewing photometric imagery.

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### **5** Summary and conclusions

- The new PARASOL/GRASP products were extensively evaluated using validations against AERONET and comparisons with the original POLDER algorithm (PARASOL/Operational), and MODIS Collection 6 aerosol products. The study was focused on the main aerosol parameters AOD, AE, AODF, AODC, SSA and AAOD included in all PARASOL/GRASP products. Level 3 data quality filtered and aggregated to 0.1 degree spatial resolution were used. The validation of PARASOL/GRASP spectral products (443 – 1020 nm) was done for the full PARASOL archive (2005-2013) against all available
- AERONET products. In addition to the direct validation of the full archive of PARASOL satellite products, the PARASOL/GRASP products were intensively inter-compared with the widely used MODIS/AQUA aerosol products from DT, DB and MAIAC (land only) algorithms and PARASOL/Operational aerosol products for one full year, 2008, at 0.1 degree (~10 km) resolution. A global comparison with AERONET for the year 2008 was performed for all products and the results inter-compared. The percentage of the cases when the product of each algorithm showed the best statistical metrics among all the products was used as an indicator for the performance evaluation. In addition, in order to further clarify the level of consistency of the satellite products, the comparisons of seasonal means as well as the global correlations of different satellite products at 0.1 degree or 0.2 degree were comprehensively analyzed for the year 2008. In terms of data volume and geographic extent, the global comparisons analyses are more representative of

1435 the global aerosol system than the subset based on colocations with AERONET.

The results show that the PARASOL/GRASP retrieval provided reliable aerosol products, and important advancement over the reference MODIS aerosol products:

- <u>Total AOD</u>

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the PARASOL spectral products including AOD for six wavelengths in the range 443 to 1020 nm agree well with AERONET AOD measurements, e.g. for <u>PARASOL/Models AOD</u> correlation coefficients R are ≥ 0.86 over land and ≥ 0.94 over ocean with BIAS not exceeding 0.01 over land and 0.02 over ocean for all wavelengths. <u>PARASOL/Optimized and PARASOL/HP also show good</u>

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spectrally;

- the AOD (550 nm) products from PARASOL/GRASP (especially GRASP/Models) correlate with AERONET generally similar or better than the correlations of MODIS AOD (550 nm) results both over ocean and land:
  - over ocean: all PARASOL (including Operational) and MODIS DT algorithms provide comparable and well correlated retrieval results;
  - over land: PARASOL/GRASP provides full land coverage products that correlate generally better with AERONET; MAIAC shows the highest percentage falling with the GCOS criteria and lowest RMSE among MODIS products, but greater overall BIAS than either DT or DB.
- the correlation between different PARASOL/GRASP products obtained only over AERONET sites and globally are rather consistent, while the correlations between PARASOL and MODIS products for global analysis over land notably degrade compared for those obtained only over AERONET sites, especially for MAIAC and DB. This finding suggests possible dependence of MODIS retrievals on AERONET regional assumptions of aerosol types or AERONET-assisted atmospheric correction to determine surface reflectance, while GRASP retrievals do not use any location specific aerosol or surface assumptions.
  - <u>AE:</u>
    - the PARASOL products agree with AERONET generally similar to the MODIS DT product over ocean and significantly better over land;
- all PARASOL/GRASP products (Optimized, HP and Models) provide AE values globally over land and ocean that agree between themselves consistently over AERONET sites and globally;

- AODF and AODC:

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- all PARASOL/GRASP products (Optimized, HP and Models) provide spectral AODF and AODC values globally over ocean and all land covers including bright surfaces, and the different products agree between themselves consistently over AERONET sites and globally;
- the PARASOL/GRASP uniquely provides AODF and AODC with global coverage;
   PARASOL/Operational provides only AODF over land, while MODIS AODF and AODC products are only available over ocean;
- the PARASOL/GRASP AODF and AODC products agree with AERONET as well as MODIS (and PARASOL/Operational) and somewhat better over ocean;

- Aerosol absorption:

- all PARASOL/GRASP products (Optimized, HP and Models) provide SSA and AAOD spectral values that are generally not accessible from MODIS and other satellite products;
- · the validation of PARASOL/GRASP shows robust correlation of the retrieved SSA and AAOD
- spectral values with AERONET (440–1020 nm), correlations increase for retrievals corresponding to the events with higher AOD. For AAOD retrievals overall the BIAS does not exceed 0.01, suggesting that PARASOL products can be used for making global estimations of AAOD at such level of uncertainty.
- Analysis presented in this paper suggests that the data from PARASOL, and therefore from multi-angle
  polarimeters (MAP) in general, allow not only solid retrievals of conventional aerosol products (e.g. AOD at 550 nm), but also detailed aerosol properties such as AOD for the whole spectrum of observations (e.g. for PARASOL from 443 to 1020 nm), and aerosol SSA and AAOD that are practically not accessible from mono- and bi- viewing photometric satellite observations, as well as improved AE, AODF, and AODC at global scale. It is also important to emphasize that PARASOL/GRASP retrievals are based on rigorous
  optimized inversion that searches for statistically optimized fitting in a continuous space of solution without using widely used Look-up-Tables. As a result, it provides a globally-consistent product using

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exactly the same aerosol modeling approach over land and ocean, unique set of a priori constraints and

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initial guess, while retrieving surface reflectance properties simultaneously with aerosol. It is expected that

- similar type of approaches will become more common and evolve further in the coming era of multiple MAP instruments, e.g. 3MI, DPC, Aerosol-UA, SPEXone and HARP2, etc. (see more in Dubovik et al., 2019). The multi-dimensional aerosol information derived from MAPs is expected to improve quality and utility of atmospheric aerosol characterization from space.
- One key finding of this work is that the best retrieval of total AOD is provided by the GRASP/Models approach, which restrains the retrieval to a priori aerosol model components, vastly 1500 reducing the number of free parameters for retrieval. The more complex GRASP/HP retrieval with many more retrieval parameters seemed to offer more accurate detailed aerosol parameters such as AE, AODF, AODC and SSA. Future efforts on improving the GRASP retrieval will be aimed at achieving accurate retrievals within one approach. However, this situation also reveals the challenge of a developing unique approach that can provide a retrieval of all parameters with highest accuracy from MAP observations. 1505 Indeed, multi-angular polarimetric observations have sensitivity to different aerosol properties, and therefore the MAP algorithms tend to be designed for the retrieval of large number of parameters, while in the situations with low aerosol presence the information may be not sufficient to retrieve all parameters reliably. Nonetheless, the presented results demonstrate an overall clear advantages of MAP aerosol retrievals compare with photometric mono-viewing product and support high expectations from future 1510 MAP missions with improved instrumental and algorithmic developments.

### Data availability

The PARASOL/GRASP Optimized, HP and Models products are publicly available the official GRASP algorithm website (<u>https://www.grasp-open.com/products</u>) and the AERIS/ICARE Data and Services 1515 Center (<u>http://www.icare.univ-lille.fr</u>). The dataset used in the current study is registered under: http://doi.org/10.5281/zenodo.3887265 (Chen et al., 2020).

### Author contribution

The GRASP aerosol products evaluation exercise has been implemented and investigated by the GRASP team (OD, DF, PL, TL AL, CC). CC and OD carried out this study and analysis. The results were widely

1520 discussed with POLDER/Operational and MODIS (DT, DB and MAIAC) aerosol team, who are co-authors of this paper. CC and OD wrote the manuscript with contributions from all authors. FD, JD, MA, LB, DM, AH and CF carried through the POLDER data processing based on the GRASP-OPEN software.

### **Competing interests**

The authors declare that they have no conflict of interest.

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# Table 1. List of aerosol parameters in PARASOL/GRASP products

Parameters	Dimension	Description
AOD	(λ, Latitude, Longitude)	Aerosol optical depth
AExp	(Latitude, Longitude)	AE (670/865 nm)
AODF	$(\lambda, Latitude, Longitude)$	Fine mode AOD
AODC	$(\lambda, Latitude, Longitude)$	Coarse mode AOD
AAOD	$(\lambda, Latitude, Longitude)$	Aerosol absorption optical depth
SSA	$(\lambda, Latitude, Longitude)$	Single scattering albedo
RealRefIndSpect	$(\lambda, Latitude, Longitude)$	Real part of refractive index
ImagRefIndSpect	$(\lambda, Latitude, Longitude)$	Imaginary part of refractive index
SizeDistrLogNormBin	(5, Latitude, Longitude)	5 Bins of size distribution
SphereFraction	(Latitude, Longitude)	Sphere fraction
VertProfileHeight	(Latitude, Longitude)	Aerosol scale height (unit: m)
LandPercentage	(Latitude, Longitude)	Land percentage (%)
ResidualRelative	(Latitude, Longitude)	Relative residual

 $\lambda = 443, 490, 565, 670, 865, and 1020 \text{ nm}$ 

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# Table 2. Strategies used to select quality assured PARASOL and MODIS aerosol products

		1	
		Land	Ocean
POLDER	GRASP/Optimized	"ResidualRelative"<0.05	"ResidualRelative"<0.1
	GRASP/HP	"ResidualRelative"<0.05	"ResidualRelative"<0.1
	GRASP/Models	"ResidualRelative"<1.0	"ResidualRelative"<1.0
	Operational	0.8≤Quality Index≤1.0	0.8≤Quality Index≤1.0
MODIS	DT	QA Flag = 3	QA Flag = 3
	DB	QA Flag = 3	_1
	MAIAC	QA = '0000'	_2

<sup>1</sup>DB aerosol product is not available over ocean.

<sup>2</sup> MAIAC aerosol product is presently only available for tiles containing land, so the ocean retrievals are not

1975 considered in this study.

approaches by each metric is labelled in bold.

Land/Ocean	Band	Products	R	Slope	Offset	RMSE	GCOS (%)	BIAS	BIAS	BIAS	BIAS
	(nm)								τ<0.2	$0.2 \leq \tau \leq 0.7$	$\tau > 0.7$
Land	443	Optimized (41268)	0.900	0.867	0.104	0.179	26.7	0.06	0.09	0.06	-0.06
		HP (42202)	0.915	0.981	0.072	0.181	32.7	0.07	0.07	0.07	0.05
		Models (28449)	0.932	1.013	0.003	0.140	49.3	0.01	0.01	0.00	0.02
	490	Optimized (41268)	0.892	0.879	0.099	0.171	26.8	0.06	0.08	0.06	-0.04
		HP (42202)	0.909	1.000	0.069	0.174	33.2	0.07	0.07	0.07	0.07
		Models (28449)	0.929	1.025	0.003	0.131	51.6	0.01	0.01	0.01	0.03
	550	Optimized (41268)	0.876	0.847	0.101	0.162	27.5	0.06	0.08	0.05	-0.08
		HP (42202)	0.898	0.973	0.074	0.163	34.0	0.07	0.07	0.07	0.04
		Models (28449)	0.922	1.023	0.005	0.123	54.2	0.01	0.01	0.01	0.03
	565	Optimized (41268)	0.877	0.877	0.096	0.161	27.3	0.06	0.08	0.06	-0.05
		HP (42202)	0.898	1.004	0.069	0.165	34.0	0.07	0.07	0.07	0.07
		Models (28449)	0.920	1.011	0.006	0.120	54.4	0.01	0.01	0.00	0.02
	670	Optimized (41268)	0.858	0.823	0.099	0.152	28.4	0.06	0.08	0.05	-0.10
		HP (42202)	0.886	0.955	0.077	0.153	35.0	0.07	0.07	0.07	0.02
		Models (28449)	0.911	0.954	0.016	0.108	58.6	0.01	0.01	-0.01	-0.03
	865	Optimized (41268)	0.816	0.785	0.093	0.142	31.3	0.05	0.07	0.03	-0.15
		HP (42202)	0.856	0.932	0.074	0.142	37.6	0.06	0.06	0.07	-0.02
		Models (284449)	0.880	0.935	0.018	0.105	60.3	0.01	0.02	-0.01	-0.04
	1020	Optimized (40148)	0.791	0.772	0.089	0.139	32.8	0.05	0.07	0.02	-0.17
		HP (41016)	0.837	0.924	0.073	0.138	38.8	0.06	0.06	0.06	-0.03
		Models (27551)	0.856	0.943	0.023	0.109	59.5	0.01	0.02	0.00	-0.04
Ocean	443	Optimized (1495)	0.938	1.028	0.049	0.084	40.5	0.05	0.05	0.07	0.03
		HP (1551)	0.939	1.043	0.046	0.083	41.2	0.05	0.05	0.06	0.05
		Models (2064)	0.940	0.970	0.026	0.066	60.6	0.02	0.02	0.03	-0.06
	490	Optimized (1495)	0.939	1.064	0.041	0.079	43.2	0.05	0.04	0.07	0.05
		HP (1551)	0.942	1.077	0.039	0.079	43.1	0.05	0.05	0.07	0.09
		Models (2064)	0.946	0.969	0.023	0.057	65.1	0.02	0.02	0.02	-0.05
	550	Optimized (1495)	0.936	1.060	0.035	0.071	48.4	0.05	0.04	0.06	0.04
		HP (1551)	0.940	1.083	0.036	0.074	46.4	0.05	0.04	0.07	0.11
		Models (2064)	0.950	0.960	0.019	0.050	70.3	0.01	0.01	0.01	-0.05
	565	Optimized (1495)	0.939	1.090	0.033	0.072	48.5	0.05	0.04	0.07	0.05
		HP (1551)	0.943	1.105	0.033	0.074	46.7	0.05	0.04	0.07	0.12
		Models $(2064)$	0.950	0.939	0.020	0.048	71.2	0.01	0.01	0.00	-0.07
		1000013 (2004)	0.200								
	670	Optimized (1495)	0.936	1.071	0.030	0.064	55.8	0.04	0.04	0.06	0.02
	670	Optimized (1495) HP (1551)	0.936 0.943	<b>1.071</b> 1.099	0.030 0.032	0.064 0.068	55.8 50.9	0.04 0.05	0.04 0.04	0.06 0.07	<b>0.02</b> 0.11

865	Optimized (1495)	0.931	1.077	0.020	0.053	66.0	0.03	0.03	0.05	0.15
	HP (1551)	0.942	1.129	0.024	0.060	58.3	0.04	0.03	0.06	0.17
	Models (2064)	0.955	0.852	0.015	0.038	82.1	0.00	0.00	-0.03	-0.13
1020	Optimized (1431)	0.927	1.063	0.017	0.049	71.3	0.02	0.02	0.04	0.15
	HP (1501)	0.940	1.143	0.021	0.058	60.9	0.04	0.03	0.07	0.18
	Models (2002)	0.957	0.865	0.013	0.035	84.6	0.00	0.00	-0.03	-0.11

# Table 4. Global statistics of PARASOL/GRASP AE vs. AERONET AE (440/870) over land and ocean.

# 1980 The best performing of three approaches by each metric is labelled in bold.

		R	Slope	Offset	RMSE	BIAS	
Land	Optimized (18594)	0.797	0.680	0.213	0.358	-0.10	
	HP (19093)	0.843	0.716	0.139	0.336	-0.14	
	Models (11468)	0.681	0.415	0.511	0.420	-0.04	
Ocean	Optimized (363)	0.935	0.773	0.199	0.210	0.01	
	HP (391)	0.949	0.817	0.092	0.193	-0.05	
	Models (522)	0.958	0.620	0.451	0.292	0.16	

		R	Slope	Offset	RMSE	GCOS	BIAS	BIAS	BIAS	BIAS
						Fraction (%)		$\tau_{f(550)} < 0.2$	$0.2 {\leq}  \tau_{f(550)} {\leq} 0.7$	$\tau_{f(550)} > 0.7$
Land	Optimized (31902)	0.922	0.840	0.044	0.100	54.9	0.02	0.03	0.01	-0.16
	HP (32973)	0.924	0.892	0.029	0.098	60.9	0.01	0.02	0.01	-0.10
	Models (23653)	0.868	0.662	0.028	0.094	64.5	-0.02	0.00	-0.07	-0.37
Ocean	Optimized (1074)	0.901	0.958	0.042	0.058	56.7	0.04	0.04	0.05	-0.24
	HP (1155)	0.908	0.932	0.028	0.043	76.3	0.02	0.02	0.03	-0.27
	Models (1338)	0.834	0.746	0.035	0.048	77.5	0.02	0.02	-0.03	-0.33

### of three approaches by each metric is labelled in bold.

### 85 Table 6. Global statistics of PARASOL/GRASP AODC vs. AERONET SDA AODC at 550 nm over land and ocean. The best performing

# of three approaches by each metric is labelled in bold.

		R	Slope	Offset	RMSE	GCOS	BIAS	BIAS	BIAS	BIAS
						Fraction (%)		$\tau_{c(550)} < 0.2$	$0.2 {\leq} \tau_{c(550)} {\leq} 0.7$	$\tau_{c (550)} > 0.7$
Land	Optimized (31903)	0.686	0.744	0.062	0.117	43.1	0.04	0.05	-0.03	-0.18
	HP (32973)	0.742	0.933	0.057	0.124	47.7	0.05	0.05	0.03	-0.02
	Models (23651)	0.571	0.653	0.040	0.112	64.3	0.02	0.03	-0.08	-0.28
Ocean	Optimized (1076)	0.871	0.942	0.009	0.046	77.6	0.01	0.00	0.01	0.03
	HP (1156)	0.915	1.119	0.015	0.051	70.0	0.02	0.02	0.06	0.20
	Models (1337)	0.922	0.754	0.010	0.036	84.8	-0.01	0.00	-0.06	-0.09

# 1990 PARASOL AOD (565 nm) levels. The best performing at each wavelength of three approaches by

# each metric is labelled in bold.

AOD Level	Band (nm)	Products	R	Slope	Offset	RMSE	BIAS
All L3 data	443	Optimized (7192)	0.285	0.292	0.631	0.051	-0.01
		HP (7450)	0.254	0.266	0.666	0.051	0.00
		Models (6095)	0.348	0.349	0.582	0.047	-0.01
	670	Optimized (7192)	0.511	0.608	0.324	0.065	-0.04
		HP (7450)	0.536	0.648	0.299	0.056	-0.03
		Models (6095)	0.321	0.334	0.602	0.057	-0.02
	865	Optimized (7192)	0.566	0.667	0.267	0.068	-0.04
		HP (7450)	0.594	0.698	0.253	0.058	-0.03
		Models (6095)	0.360	0.347	0.597	0.059	-0.01
	1020	Optimized (7192)	0.596	0.705	0.230	0.072	-0.04
		HP (7450)	0.627	0.730	0.223	0.060	-0.03
		Models (6095)	0.372	0.334	0.615	0.062	0.00
AOD>0.5	443	Optimized (3695)	0.315	0.312	0.619	0.045	-0.01
		HP (4235)	0.242	0.242	0.691	0.047	0.00
		Models (2424)	0.413	0.345	0.594	0.037	0.00
	670	Optimized (3695)	0.534	0.612	0.327	0.056	-0.04
		HP (4235)	0.552	0.642	0.307	0.051	-0.03
		Models (2424)	0.455	0.355	0.592	0.042	-0.01
	865	Optimized (3695)	0.593	0.668	0.274	0.059	-0.04
		HP (4235)	0.615	0.699	0.254	0.052	-0.03
		Models (2424)	0.535	0.387	0.571	0.043	0.00
	1020	Optimized (3695)	0.627	0.703	0.240	0.061	-0.04
		HP (4235)	0.647	0.726	0.228	0.054	-0.03
		Models (2424)	0.564	0.376	0.586	0.046	0.00
AOD>1.0	443	Optimized (715)	0.478	0.459	0.499	0.034	0.00
		HP (976)	0.398	0.366	0.587	0.037	0.00
		Models (463)	0.585	0.457	0.499	0.027	0.01
	670	Optimized (715)	0.674	0.712	0.252	0.036	-0.02
		HP (976)	0.664	0.687	0.277	0.036	-0.02
		Models (463)	0.665	0.464	0.497	0.031	-0.01
	865	Optimized (715)	0.702	0.699	0.264	0.039	-0.02
		HP (976)	0.704	0.692	0.272	0.037	-0.02
	1020	Models (463)	0.737	0.487	0.483	0.033	0.00
	1020	Optimized (715)	0.715	0.694	0.268	0.042	-0.02
		HP(976)	0.723	0.699	0.265	0.040	-0.02
100.15	112	Models (463)	0.757	0.453	0.519	0.038	0.01
AOD>1.5	443	Optimized (212)	0.544	0.536	0.430	0.030	0.00
		HP(317)	0.527	0.518	0.459	0.031	0.00
	(70)	Models (116)	0.639	0.491	0.472	0.022	0.00
	670	Up (217)	0.734	0.752	0.220	0.030	-0.01
		HP(317)	0.752	0.804	0.171	0.029	-0.01
	0(5	Models (116)	0.814	0.567	0.402	0.023	0.00
	865	Up (217)	0.760	0.688	0.283	0.032	-0.01
		HP (317) Medela (116)	0.770	0.738	0.235	0.030	-0.01
	1020	Ontimized (212)	0.770	0.602	0.373	0.025	0.00
	1020	Up $(217)$	0.770	0.000	0.303	0.033	-0.01
		HP (317) Medela (116)	0.//9	0.710	0.423	0.034	-0.01
		wodels (116)	0.889	0.556	0.425	0.032	0.01

# Table 8. Global statistics of PARASOL/GRASP spectral AAOD vs. AERONET AAOD. The best

Band (nm)	Products	R	Slope	Offset	RMSE	BIAS	
443	Optimized (7192)	0.486	0.475	0.040	0.046	0.01	
	HP (7450)	0.498	0.536	0.034	0.047	0.00	
	Models (8046)	0.538	0.509	0.035	0.042	0.00	
670	Optimized (7192)	0.480	0.571	0.033	0.034	0.02	
	HP (7450)	0.517	0.673	0.028	0.034	0.02	
	Models (8046)	0.480	0.492	0.023	0.026	0.01	
865	Optimized (7192)	0.393	0.476	0.029	0.028	0.02	
	HP (7450)	0.438	0.574	0.024	0.028	0.01	
	Models (8046)	0.444	0.439	0.017	0.020	0.00	
1020	Optimized (7192)	0.343	0.430	0.026	0.025	0.01	
	HP (7450)	0.394	0.526	0.022	0.025	0.01	
	Models (8046)	0.414	0.409	0.015	0.018	0.00	

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performing at each wavelength of three approaches by each metric is labelled in bold.

			Refe	rence: AEI	RONET AO	$D(\tau_{550})$				
		R	Slope	Offset	RMSE	GCOS	BIAS	BIAS	BIAS	BIAS
						Fraction (%)		$\tau_{550} < 0.2$	$0.2 \le \tau_{550} \le 0.7$	$\tau_{550} > 0.7$
Land	GRASP/Optimized (3647)	0.875	0.780	0.098	0.150	28.8	0.04	0.07	0.02	-0.13
	GRASP/HP (4777)	0.908	0.938	0.078	0.157	32.4	0.06	0.07	0.06	0.02
	GRASP/Models (3111)	0.924	0.989	0.005	0.121	53.2	0.00	0.01	-0.01	0.02
	DT (6858)	0.898	0.988	0.021	0.120	46.1	0.02	0.02	0.02	0.00
	DB (8409)	0.870	0.841	0.026	0.126	48.8	-0.01	0.01	-0.04	-0.14
	MAIAC_0.1 (8164)	0.895	0.793	0.007	0.112	52.8	-0.03	-0.01	-0.08	-0.19
	MAIAC_0.01 (9054)	0.874	0.796	0.014	0.125	48.1	-0.03	0.00	-0.08	-0.19
Ocean	GRASP/Optimized (116)	0.950	1.145	0.033	0.089	42.4	0.06	0.05	0.09	0.17
	GRASP/HP (154)	0.947	1.074	0.054	0.092	26.6	0.07	0.06	0.08	0.13
	GRASP/Models (205)	0.963	0.965	0.024	0.061	62.9	0.02	0.02	0.02	-0.04
	Operational (207)	0.954	1.165	-0.009	0.077	52.2	0.03	0.01	0.04	0.18
	DT (218)	0.952	0.974	0.037	0.081	55.0	0.03	0.03	0.04	0.00

Table 9. Statistics of PARASOL and MODIS AOD products against collocated AERONET AOD at 550 nm over land and ocean

Table 10. Statistics of PARASOL and MODIS AOD products against collocated AERONET AOD at 550 nm classified by NDVI. In each

individual NDVI level, the best performing metric is indicated in bold. The number of matched pairs is included in brackets.

Reference: AERONET AOD ( $\tau_{550}$ )											
		R	Slope	Offset	RMSE	GCOS	BIAS	BIAS	BIAS	BIAS	
						Fraction (%)		$\tau_{550} < 0.2$	$0.2 \le \tau_{550} \le 0.7$	$\tau_{550} > 0.7$	
Land	GRASP/Optimized (1055)	0.892	0.711	0.105	0.153	23.0	0.02	0.08	-0.01	-0.20	
NDVI<0.2	GRASP/HP (1410)	0.915	0.860	0.104	0.155	26.0	0.06	0.09	0.05	-0.03	
	GRASP/Models (786)	0.873	0.888	0.023	0.159	39.8	-0.01	0.01	-0.03	0.01	
	DT (99)	0.792	0.878	0.073	0.216	44.4	0.05	0.04	0.12	-0.04	
	DB (1327)	0.845	0.790	0.044	0.153	44.2	-0.01	0.03	-0.05	-0.16	
	MAIAC_0.1 (1853)	0.883	0.734	0.032	0.139	42.4	-0.03	0.02	-0.09	-0.22	
	MAIAC_0.01 (2087)	0.853	0.734	0.041	0.155	35.7	-0.03	0.03	-0.09	-0.22	
Land	GRASP/Optimized (1106)	0.881	0.777	0.101	0.161	31.9	0.04	0.07	0.03	-0.16	
0.2≤NDVI<0.4	GRASP/HP (1479)	0.928	0.911	0.074	0.145	39.4	0.05	0.05	0.06	-0.03	
	GRASP/Models (1020)	0.953	1.062	-0.014	0.125	52.7	0.00	0.00	-0.01	0.04	
	DT (1847)	0.895	0.947	0.029	0.145	40.6	0.02	0.03	0.01	-0.05	
	DB (2204)	0.888	0.883	0.010	0.142	46.3	-0.02	0.00	-0.05	-0.09	
	MAIAC_0.1 (2049)	0.901	0.825	-0.005	0.133	53.9	-0.04	-0.01	-0.11	-0.18	
	MAIAC_0.01 (2363)	0.896	0.826	0.002	0.134	51.5	-0.04	0.00	-0.10	-0.18	Cheng Chen 3/11/2020 14:24
Land	GRASP/Optimized (958)	0.880	0.868	0.083	0.138	31.7	0.05	0.07	0.03	-0.02	Formatted: Font:Bold, English (US)
0.4≤NDVI<0.6	GRASP/HP (1249)	0.903	1.069	0.047	0.173	33.2	0.07	0.06	0.07	0.12	
	GRASP/Models (1074)	0.920	0.952	0.014	0.086	61.5	0.00	0.01	0.00	-0.04	
	DT (2702)	0.907	0.994	0.012	0.112	46.6	0.01	0.01	0.02	-0.01	
	DB (2718)	0.866	0.808	0.030	0.120	50.0	-0.01	0.01	-0.03	-0.20	
	MAIAC_0.1 (2193)	0.911	0.821	-0.009	0.093	53.5	-0.04	-0.03	-0.07	-0.19	
	MAIAC_0.01 (2530)	0.899	0.827	-0.002	0.097	50.5	-0.03	-0.02	-0.07	-0.17	_
Land	GRASP/Optimized (194)	0.832	0.932	0.108	0.145	23.7	0.09	0.10	0.08	0.09	
NDVI≥0.6	GRASP/HP (287)	0.853	1.001	0.107	0.160	21.3	0.11	0.10	0.11	0.10	
	GRASP/Models (231)	0.910	1.115	0.011	0.083	61.9	0.03	0.02	0.07	0.06	
	DT (943)	0.910	1.118	-0.005	0.076	55.0	0.01	0.01	0.03	0.17	
	DB (907)	0.855	0.884	0.015	0.076	60.2	0.00	0.01	-0.04	0.07	
	MAIAC_0.1 (651)	0.826	0.837	-0.005	0.063	66.2	-0.02	-0.02	-0.06	-0.04	
	MAIAC_0.01 (669)	0.840	0.929	-0.013	0.074	61.3	-0.02	-0.02	-0.04	0.10	_

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	Reference: AERONET AE										
		R	Slope	Offset	RMSE	BIAS					
Land	GRASP/Optimized (2035)	0.745	0.641	0.168	0.435	-0.19					
	GRASP/HP (2791)	0.772	0.654	0.122	0.425	-0.21					
	GRASP/Models (1253)	0.686	0.407	0.507	0.443	-0.06					
	DT (2589)	0.390	0.372	0.514	0.599	-0.31					
	DB (3279)	0.563	0.650	0.444	0.573	0.04					
Ocean	GRASP/Optimized (55)	0.840	0.724	0.183	0.279	-0.02					
	GRASP/HP (80)	0.890	0.810	0.051	0.229	-0.08					
	GRASP/Models (92)	0.949	0.625	0.431	0.291	0.20					
	Operational (57)	0.891	0.812	0.841	0.782	0.75					
	DT (106)	0.832	0.610	0.317	0.305	0.08					

land and ocean, with a threshold of satellite AOD (550 nm) > 0.2.

	Reference: AERONET AODF ( $\tau_{f 550}$ )											
		R	Slope	Offset	RMSE	GCOS	BIAS	BIAS	BIAS	BIAS		
						Fraction (%)		$\tau_{f(550)} < 0.2$	$0.2 {\leq} \tau_{f(550)} {\leq} 0.7$	$\tau_{f(550)} > 0.7$		
Land	GRASP/Optimized (2634)	0.923	0.762	0.043	0.104	58.0	0.00	0.02	-0.01	-0.23		
	GRASP/HP (3507)	0.926	0.828	0.036	0.106	59.5	0.01	0.02	0.00	-0.17		
	GRASP/Models (2795)	0.868	0.587	0.035	0.124	63.8	-0.02	0.00	-0.08	-0.46		
	Operational (2619)	0.886	0.546	0.052	0.162	50.5	-0.04	0.00	-0.08	-0.45		
Ocean	GRASP/Optimized (91)	0.893	1.397	0.023	0.079	40.7	0.06	0.05	0.10			
	GRASP/HP (129)	0.924	1.118	0.018	0.049	75.2	0.03	0.03	0.05			
	GRASP/Models (168)	0.866	1.046	0.028	0.054	65.5	0.03	0.03	0.02			
	Operational (82)	0.780	1.082	0.017	0.061	67.1	0.02	0.02	0.00			
	DT (119)	0.808	0.887	0.048	0.067	56.3	0.04	0.04	-0.01			

Table 12. Statistics of PARASOL and MODIS AODF products against collocated AERONET AODF over land and ocean

# 05 Table 13. Statistics of PARASOL and MODIS AODC products against collocated AERONET AODC over land and ocean

Reference: AERONET AODC ( $\tau_{c 550}$ )											
		R	Slope	Offset	RMSE	GCOS	BIAS	BIAS	BIAS	BIAS	
						Fraction (%)		$\tau_{c(550)} < 0.2$	$0.2 \le \tau_{c(550)} \le 0.7$	$\tau_{c(550)} > 0.7$	
Land	GRASP/Optimized (2634)	0.700	0.678	0.058	0.114	45.6	0.03	0.04	-0.06	-0.24	
	GRASP/HP (3506)	0.771	0.912	0.060	0.127	45.8	0.05	0.06	0.02	0.00	
	GRASP/Models (2795)	0.536	0.596	0.043	0.125	63.7	0.01	0.03	-0.12	-0.28 •	
Ocean	GRASP/Optimized (91)	0.936	1.033	0.021	0.062	59.3	0.03	0.02	0.05	0.03	
	GRASP/HP (129)	0.961	1.113	0.033	0.070	45.0	0.05	0.04	0.07	0.20	
	GRASP/Models (168)	0.966	0.827	0.008	0.040	81.5	-0.01	0.00	-0.05	-0.09	
	Operational (82)	0.936	0.971	0.014	0.045	74.4	0.01	0.01	-0.01		
	DT (119)	0.911	0.806	0.025	0.045	68.9	0.00	0.01	-0.03	-0.11	

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	Reference: PARASOL/Operational											
			R	Slope	Offset	RMSE	BIAS	BIAS	BIAS	BIAS		
								$\tau/\tau_f/\tau_c < 0.2$	$0.2 \le \tau/\tau_f/\tau_c \le 0.7$	$\tau/\tau_f/\tau_c > 0.7$		
Land	AODF	GRASP/HP	0.88 (0.91)	1.22 (1.13)	0.03 (0.02)	0.13 (0.13)	0.06 (0.05)	0.04 (0.04)	0.10 (0.05)	0.29 (0.18)		
		8 209 015 (7801)										
		GRASP/Models	0.85 (0.83)	0.94 (0.74)	0.03 (0.04)	0.10 (0.13)	0.02 (-0.01)	0.03 (0.01)	0.02 (-0.04)	-0.05 (-0.19)		
		8 117 246 (7427)										
Ocean	AOD	GRASP/HP	0.88 (0.98)	1.19 (1.22)	0.04 (0.04)	0.10 (0.16)	0.06 (0.11)	0.06 (0.08)	0.10 (0.10)	0.22 (0.38)		
		5 702 109 (93)										
		GRASP/Models	0.94 (0.99)	1.09 (1.14)	0.00 (0.01)	0.04 (0.09)	0.01 (0.05)	0.01 (0.04)	0.03 (0.03)	0.10 (0.24)		
		5 988 842 (91)										
	AE	GRASP/HP	0.59 (0.67)	0.53 (0.62)	-0.04 (-0.18)	0.62 (0.77)	-0.48 (-0.66)					
		5 703 282 (93)										
		GRASP/Models	0.38 (0.53)	0.24 (0.40)	0.69 (0.41)	0.44 (0.53)	-0.03 (-0.34)					
		5 988 842 (91)										
	AODF	GRASP/HP	0.63 (0.41)	0.52 (0.24)	0.05 (0.12)	0.07 (0.15)	0.02 (0.00)	0.03 (0.05)	-0.12 (-0.09)	-0.40 (-0.64)		
		5 704 665 (126)										
		GRASP/Models	0.73 (0.74)	0.57 (0.58)	0.04 (0.09)	0.06 (0.11)	0.01 (0.02)	0.02 (0.05)	-0.12 (-0.06)	-0.31 (-0.23)		
		5 991 408 (125)										
	AODC	GRASP/HP	0.68 ()	1.08 ()	0.04 ()	0.10 ()	0.05 ()	0.04 ()	0.17 ()	0.53 ()		
		2 692 908 ()										
		GRASP/Models	0.70 ()	0.69 ()	0.02 ()	0.06 ()	0.00 ()	0.00 ()	-0.01 ()	0.04 ()		
		2 949 016 ()										

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Table 15. Pixel to pixel (0.1° x 0.1°) statistical metrics between AOD (550 nm) products based on references of GRASP/Models (Land

	Reference: GRASP/Models AOD										
		R	Slope	Offset	RMSE	BIAS	BIAS	BIAS	BIAS		
			-				$\tau_{550} < 0.2$	$0.2 \le \tau_{550} \le 0.7$	$\tau_{550} > 0.7$		
Land	GRASP/HP AOD	0.85 (0.90)	0.81 (0.84)	0.11 (0.12)	0.19 (0.19)	0.06 (0.06)	0.07 (0.08)	0.06 (0.06)	-0.07 (-0.02)		
	53 656 407 (8564)										
	DT AOD	0.85 (0.90)	0.84 (1.01)	0.00 (0.06)	0.15 (0.18)	-0.04 (0.06)	0.00 (0.04)	-0.07 (0.07)	-0.15 (0.07)		
	13 069 294 (3 432)										
	DB AOD	0.76 (0.89)	0.69 (0.89)	0.06 (0.03)	0.18 (0.16)	-0.01 (0.00)	0.03 (0.02)	-0.04 (-0.02)	-0.26 (-0.06)		
	36 348 953 (4 972)										
	MAIAC AOD	0.77 (0.89)	0.66 (0.84)	0.05 (0.02)	0.18 (0.15)	-0.03 (-0.02)	0.02 (0.01)	-0.08 (-0.04)	-0.29 (-0.11)		
	64 921 447 (10 830)										
Ocean	GRASP/HP AOD	0.94 (0.97)	1.10 (1.04)	0.04 (0.05)	0.09 (0.10)	0.05 (0.06)	0.05 (0.05)	0.07 (0.06)	0.16 (0.11)		
	65 551 501 (300)										
	DT AOD	0.92 (0.97)	0.88 (0.99)	0.01 (-0.01)	0.05 (0.05)	-0.01 (-0.01)	0.00 (-0.02)	-0.04 (-0.01)	-0.06 (-0.04)		
	32 486 105 (130)										
	DB AOD										
	MAIAC AOD										
				Reference:	MAIAC AOD						
Land	GRASP/HP AOD	0.81 (0.90)	0.91 (0.91)	0.10 (0.11)	0.20 (0.17)	0.08 (0.08)	0.08 (0.09)	0.10 (0.09)	0.00 (0.02)		
	54 693 580 (8 679)										
	DT AOD	0.91 (0.93)	1.05 (1.07)	0.01 (0.06)	0.10 (0.15)	0.02 (0.08)	0.01 (0.05)	0.01 (0.11)	0.07 (0.12)		
	21 272 908 (5 836)										
	DB AOD	0.86 (0.93)	0.92 (0.98)	0.04 (0.03)	0.13 (0.12)	0.02 (0.02)	0.03 (0.02)	0.02 (0.02)	-0.05 (0.00)		
	53 758 759 (7 681)										

15 and Ocean) and MAIAC AOD (Land); the statistics for only AERONET pixels are presented in brackets.

20

Table 16. Pixel to pixel (0.1° x 0.1°) statistical metrics between AOD products based on reference of GRASP/Models over land pixels with four classes of surface (NDVI<0.2, 0.2≤NDVI<0.4, 0.4≤NDVI<0.6, and NDVI≥0.6); the statistics for only AERONET pixels are presented in brackets.

Reference: GRASP/Models AOD										
		R	Slope	Offset	RMSE	BIAS	BIAS	BIAS	BIAS	
							$\tau_{550} \le 0.2$	$0.2 \le \tau_{550} \le 0.7$	$\tau_{550} > 0.7$	
Land	GRASP/HP AOD	0.78 (0.81)	0.68 (0.66)	0.15 (0.19)	0.20 (0.28)	0.06 (0.08)	0.09 (0.12)	0.07 (0.09)	-0.20 (-0.21)	
NDVI<0.2	31 341 330 (2069)									
	DT AOD	0.74 (0.88)	0.95 (1.27)	0.09 (0.16)	0.17 (0.31)	0.09 (0.24)	0.09 (0.18)	0.07 (0.26)	0.12 (0.47)	
	542 625 (38)									
	DB AOD	0.66 (0.82)	0.59 (0.74)	0.11 (0.08)	0.22 (0.21)	0.01 (0.00)	0.06 (0.05)	-0.02 (-0.03)	-0.33 (-0.17)	
	17 834 405 (1013)	0.65.00.50		0.00 (0.00)	0.01 (0.01)	0.01 ( 0.01)	0.04 (0.05)	0.0440.040	0.00 ( 0.00	
	MAIAC AOD	0.67 (0.79)	0.60 (0.68)	0.08 (0.08)	0.21 (0.21)	-0.01 (-0.01)	0.04 (0.05)	-0.06 (-0.04)	-0.32 (-0.26)	
T 1	<u>31 329 /12 (235 /)</u>	0.00 (0.02)	0.04 (0.01)	0.00 (0.00)	0.16 (0.16)	0.04 (0.04)	0.05 (0.00)	0.05 (0.05)	0.06 (0.00)	
	GRASP/HP AOD	0.90 (0.93)	0.84 (0.91)	0.08 (0.08)	0.16 (0.16)	0.04 (0.04)	0.05 (0.06)	0.05 (0.05)	-0.06 (0.00)	
$0.2 \leq NDVI \leq 0.4$	11 66 / 461 (3596)	0.01 (0.00)	0.07 (0.00)	0.01 (0.00)	0.1((0.21)	0.02 (0.00)	0.01 (0.00)	0.0( (0.10)	0.00 (0.00)	
	DT AOD 2 784 202 (1547)	0.81 (0.89)	0.86 (0.99)	0.01 (0.09)	0.16 (0.21)	-0.02 (0.08)	0.01 (0.06)	-0.06 (0.10)	-0.08 (0.08)	
	5/64 302 (1347)	0.95 (0.02)	0.78 (0.05)	0.02 (0.02)	0.14 (0.16)	0.01 (0.00)	0.01 (0.01)	0.04 (0.00)	0.15(0.02)	
	7 767 588 (1011)	0.85 (0.92)	0.78 (0.93)	0.03 (0.02)	0.14 (0.10)	-0.01 (0.00)	0.01 (0.01)	-0.04 (0.00)	-0.13 (-0.03)	
	MALAC AOD	0.87 (0.92)	0.69 (0.89)	0.04 (0.02)	0.15 (0.15)	-0.03 (-0.02)	0.01 (0.01)	-0.08 (-0.04)	-0.26 (-0.08)	
	13 927 469 (4133)	0.07 (0.92)	0.09 (0.09)	0.04 (0.02)	0.15 (0.15)	0.05 ( 0.02)	0.01 (0.01)	0.00 ( 0.04)	0.20 ( 0.00)	
Land	GRASP/HP AOD	0.92 (0.93)	0.98 (0.95)	0.07 (0.08)	0.16 (0.14)	0.06 (0.06)	0.06 (0.07)	0.05 (0.06)	0.06 (0.06)	
0.4 <ndvi<0.6< td=""><td>7 879 243 (2641)</td><td>(()))</td><td></td><td></td><td></td><td></td><td></td><td></td><td></td></ndvi<0.6<>	7 879 243 (2641)	(()))								
	DT AOD	0.86 (0.91)	0.83 (1.01)	-0.01 (0.04)	0.16 (0.14)	-0.06 (0.04)	-0.02 (0.04)	-0.08 (0.05)	-0.17 (0.05)	
	5 431 789 (1605)	, , , , , , , , , , , , , , , , , , ,	× ,	, , , , , , , , , , , , , , , , , , ,	× ,	× ,	· · · ·	~ /	× /	
	DB AOD	0.88 (0.91)	0.80 (0.89)	0.01 (0.02)	0.13 (0.13)	-0.03 (-0.01)	0.00 (0.01)	-0.07 (-0.02)	-0.16 (-0.06)	
	7 146 072 (1763)									
	MAIAC AOD	0.88 (0.92)	0.74 (0.90)	0.01 (0.00)	0.14 (0.11)	-0.05 (-0.03)	-0.01 (-0.01)	-0.10 (-0.05)	-0.24 (-0.08)	
	12 624 553 (3660)									
Land	GRASP/HP AOD	0.94 (0.89)	1.00 (0.90)	0.10 (0.10)	0.19 (0.13)	0.10 (0.08)	0.08 (0.09)	0.11 (0.07)	0.13 (0.01)	
NDVI≥0.6	2 766 521 (258)									
	DT AOD	0.91 (0.89)	0.86 (0.85)	-0.01 (0.01)	0.13 (0.09)	-0.05 (-0.02)	-0.02 (0.00)	-0.08 (-0.04)	-0.17 (-0.15)	
	3 305 544 (242)									
	DB AOD	0.90 (0.77)	0.72 (0.55)	0.00 (0.02)	0.12 (0.10)	-0.05 (-0.05)	-0.02 (-0.02)	-0.11 (-0.12)	-0.25 (-0.28)	
	3 598 331 (285)									
	MAIAC AOD	0.90 (0.85)	0.73 (0.71)	0.00 (0.00)	0.14 (0.10)	-0.06 (-0.05)	-0.02 (-0.03)	-0.11 (-0.10)	-0.28 (-0.30)	
	7 029 548 (680)									

# Table 17. The same as Table 16, but for reference of MAIAC AOD

Reference: MAIAC AOD										
		R	Slope	Offset	RMSE	BIAS	BIAS	BIAS	BIAS	
							$\tau_{550} < 0.2$	$0.2 \le \tau_{550} \le 0.7$	$\tau_{550} > 0.7$	
Land	GRASP/HP AOD	0.78 (0.86)	0.80 (0.83)	0.12 (0.13)	0.20 (0.18)	0.08 (0.08)	0.08 (0.09)	0.08 (0.10)	-0.11 (-0.08)	
NDVI<0.2	32 768 635 (2207)									
	DT AOD	0.77 (0.71)	1.28 (1.43)	0.05 (0.33)	0.15 (0.51)	0.08 (0.42)	0.07 (0.34)	0.14 (0.56)	0.33 (0.72)	
	885 841 (83)									
	DB AOD	0.85 (0.89)	0.86 (0.96)	0.06 (0.02)	0.15 (0.16)	0.04 (0.01)	0.04 (0.02)	0.03 (-0.01)	-0.09 (0.00)	
	26 151 234 (1500)									
Land	GRASP/HP AOD	0.85 (0.91)	1.03 (0.92)	0.06 (0.09)	0.17 (0.17)	0.07 (0.07)	0.06 (0.07)	0.09 (0.08)	0.08 (0.02)	
$0.2 \leq NDVI \leq 0.4$	11 919 986 (3641)									
	DT AOD	0.88 (0.94)	1.03 (1.05)	0.00 (0.08)	0.11 (0.17)	0.01 (0.10)	0.01 (0.06)	0.00 (0.13)	0.08 (0.13)	
	5 857 865 (2314)	0.07 (0.04)	1.00 (1.00)	0.01 (0.02)	0.10 (0.10)	0.01 (0.00)	0.01 (0.02)		0.05 (0.01)	
	DB AOD	0.87 (0.94)	1.02 (1.00)	0.01 (0.02)	0.12 (0.13)	0.01 (0.02)	0.01 (0.02)	0.02 (0.04)	0.05 (0.01)	
<b>T</b> 1	<u>11 668 355 (2922)</u>	0.07 (0.01)	1.00 (0.00)	0.00 (0.11)	0.01 (0.17)	0.11 (0.10)	0.00 (0.10)	0.14 (0.10)	0.17(0.00)	
Land	GRASP/HP AOD	0.87 (0.91)	1.09 (0.96)	0.09 (0.11)	0.21 (0.17)	0.11 (0.10)	0.09 (0.10)	0.14 (0.10)	0.17 (0.08)	
$0.4 \leq NDVI \leq 0.6$	/ 489 541 (2526)	0.02 (0.04)	1.02 (1.00)	0.00 (0.05)	0.10 (0.12)	0.01 (0.07)	0.01 (0.05)	0.00 (0.00)	0.05 (0.12)	
	DI AOD 9 401 721 (2410)	0.92 (0.94)	1.03 (1.06)	0.00 (0.05)	0.10 (0.13)	0.01 (0.07)	0.01 (0.05)	0.00 (0.09)	0.05 (0.12)	
	8 401 /31 (2410) DR AOD	0.88 (0.05)	0.07(0.05)	0.02 (0.04)	0.11 (0.00)	0.02 (0.03)	0.02 (0.03)	0.01 (0.02)	0.00(0.00)	
	10 298 915 (2628)	0.88 (0.95)	0.97 (0.93)	0.02 (0.04)	0.11 (0.09)	0.02 (0.03)	0.02 (0.03)	0.01 (0.03)	0.00 (0.00)	
Land	GPASP/HPAOD	0.88 (0.84)	1 13 ( 04)	0.14 (0.15)	0.26 (0.18)	0.17 (0.14)	0.13 (0.14)	0.24 (0.13)	0.26 (0.08)	
NDVI>0.6	2 512 741 (305)	0.00 (0.04)	1.15 (.)4)	0.14 (0.15)	0.20 (0.10)	0.17 (0.14)	0.13 (0.14)	0.24 (0.15)	0.20 (0.00)	
	DT AOD	0.94(0.94)	1 10 (1 06)	0.00(0.02)	0.09(0.07)	0.02 (0.03)	0.01 (0.02)	0.04 (0.05)	0.09(0.05)	
	5 539 285 (548)	0.51 (0.54)	1.10 (1.00)	0.00 (0.02)	0.07 (0.07)	0.02 (0.05)	0.01 (0.02)	0.01 (0.05)	0.07 (0.05)	
	DB AOD	0.88 (0.78)	0.91 (0.73)	0.02 (0.03)	0.09(0.06)	0.01 (0.00)	0.01 (0.01)	-0.02 (-0.05)	-0.07 (-0.27)	
	5 253 920 (520)				(0.00)		(5.01)			
	()		1		1		1			

# 35 Table 18. Pixel to pixel statistical metrics between AE products based on references of GRASP/HP; the statistics for only AERONET

pixels are presented in brackets.

	Reference: GRASP/HP AE											
		R	Slope	Offset	RMSE	BIAS						
Land	GRASP/Models AE	0.70 (0.68)	0.51 (0.43)	0.45 (0.47)	0.39 (0.39)	0.12 (-0.05)						
	27 385 356 (5 517)											
	DT AE	0.31 (0.30)	0.32 (0.29)	0.84 (0.64)	0.66 (0.59)	0.11 (-0.15)						
	6 017 122 (2 335)											
	DB AE	0.40 (0.43)	0.53 (0.49)	0.39 (0.68)	0.67 (0.65)	0.09 (0.21)						
	19 317 232 (3 121)											
Ocean	GRASP/Models AE	0.74 (0.88)	0.52 (0.68)	0.63 (0.47)	0.45 (0.33)	0.35 (0.23)						
	49 987 062 (285)											
	DT AE	0.46 (0.55)	0.49 (0.78)	0.55 (0.82)	0.53 (0.83)	0.25 (0.60)						
	18 564 876 (123)											

82

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Table 19. Pixel to pixel statistical metrics between AE products based on references of GRASP/HP over land pixels with four classes of

surface (NDVI<0.2, 0.2≤NDVI<0.4, 0.4≤NDVI<0.6, and NDVI≥0.6)

Reference: GRASP/HP AE								
		R	Slope	Offset	RMSE	BIAS		
Land	GRASP/Models AE	0.40 (0.53)	0.38 (0.48)	0.49 (0.48)	0.42 (0.42)	0.23 (0.24)		
NDVI<0.2	15 916 616 (1205)							
	DT AE	0.16 (0.36)	0.14 (0.15)	0.71 (0.47)	0.65 (0.48)	0.32 (-0.30)		
	203 121 (25)							
	DB AE	0.12 (0.35)	0.21 (0.60)	0.37 (0.40)	0.65 (0.65)	0.02 (0.21)		
	12 223 721 (764)							
Land	GRASP/Models AE	0.79 (0.69)	0.54 (0.41)	0.42 (0.47)	0.35 (0.39)	0.05 (-0.11)		
0.2 ≤ NDVI < 0.4	5 220 459 (2425)							
	DT AE	0.30 (0.33)	0.30 (0.27)	0.79 (0.58)	0.69 (0.59)	0.16 (-0.20)		
	1 923 619 (1168)							
	DB AE	0.21 (0.24)	0.23 (0.26)	0.86 (0.98)	0.77 (0.71)	0.24 (0.24)		
	3 157 768 (1256)							
Land	GRASP/Models AE	0.80 (0.65)	0.57 (0.48)	0.38 (0.43)	0.34 (0.37)	-0.11 (-0.16)		
0.4≤NDVI<0.6	4 516 281 (1743)							
	DT AE	0.28 (0.26)	0.30 (0.29)	0.90 (0.71)	0.65 (0.58)	0.08 (-0.12)		
	2 723 494 (1024)							
	DB AE	0.23 (0.27)	0.21 (0.30)	1.04 (1.00)	0.64 (0.58)	0.15 (0.19)		
	2 896 017 (999)							
Land	GRASP/Models AE	0.76 (0.73)	0.57 (0.67)	0.41 (0.24)	0.31 (0.29)	-0.08 (-0.14)		
NDVI≥0.6	1 730 292 (144)							
	DT AE	0.19 (-0.01)	0.22 (-0.01)	1.00 (1.30)	0.65 (0.60)	0.09 (0.14)		
	1 166 000 (118)							
	DB AE	0.18 (-0.07)	0.16 (-0.11)	1.21 (1.44)	0.59 (0.63)	0.25 (0.17)		
	1 039 192 (102)							

50

Table 20: Pixel to pixel statistical metrics between GRASP/HP AODF with other AODF products; the statistics for only AERONET

## pixels are presented in brackets.

Reference: GRASP/HP AODF								
		R	Slope	Offset	RMSE	BIAS		
Land	GRASP/Models AODF 53 656 407 (8 564)	0.87 (0.91)	0.75 (0.68)	0.02 (0.03)	0.10 (0.15)	-0.01 (-0.06)		
Ocean	GRASP/Models AODF 65 551 501 (300)	0.89 (0.67)	0.78 (0.90)	0.02 (0.03)	0.05 (0.11)	0.00 (0.01)		
	DT AODF 17 513 511 (116)	0.86 (0.70)	0.66 (0.64)	0.02 (0.04)	0.06 (0.09)	-0.02 (-0.03)		

55

## Table 21: The same as Table 20, but for AODC

Reference: GRASP/HP AODC							
		R	Slope	Offset	RMSE	BIAS	
Land	GRASP/Models AODC	0.71 (0.63)	0.65 (0.67)	0.02 (0.06)	0.16 (0.18)	-0.04 (0.00)	
	53 656 407 (8 564)						
Ocean	GRASP/Models AODC	0.89 (0.98)	0.56 (0.64)	0.00 (0.01)	0.09 (0.14)	-0.05 (-0.07)	
	65 551 501 (300)						
	DT AODC	0.84 (0.90)	0.58 (0.69)	0.01 (0.00)	0.08 (0.10)	-0.04 (-0.04)	
	17 513 511 (116)						

84

Table 22: Pixel to pixel statistical metrics between AODF products based on references of GRASP/HP over land pixels with four classes

of surface (NDVI<0.2, 0.2 <NDVI<0.4, 0.4 <NDVI<0.6, and NDVI ≥0.6); the statistics for only AERONET pixels are presented in brackets.

Reference: GRASP/HP AODF							
		R	Slope	Offset	RMSE	BIAS	
Land	GRASP/Models AODF	0.68 (0.82)	0.91 (0.84)	0.02 (0.03)	0.09 (0.12)	0.00 (0.01)	
NDVI<0.2	31 340 947 (2069)						
Land	GRASP/Models AODF	0.90 (0.93)	0.79 (0.68)	0.02 (0.03)	0.09 (0.16)	-0.02 (-0.07)	
0.2 ≤ NDVI < 0.4	11 667 461 (3596)						
Land	GRASP/Models AODF	0.93 (0.92)	0.73 (0.67)	0.01 (0.02)	0.13 (0.16)	-0.06 (-0.09)	
0.4 ≤ NDVI < 0.6	7 879 243 (2641)						
Land	GRASP/Models AODF	0.94 (0.88)	0.76 (0.71)	-0.01 (-0.01)	0.16 (0.12)	-0.09 (-0.08)	
NDVI≥0.6	2 766 521 (258)						

65

## Table 23: The same as Table 22, but for AODC

Reference: GRASP/HP AODC							
		R	Slope	Offset	RMSE	BIAS	
Land	GRASP/Models AODC	0.69 (0.67)	0.64 (0.72)	0.01 (-0.01)	0.18 (0.23)	-0.07 (-0.08)	
NDVI<0.2	31 340 947 (2069)						
Land	GRASP/Models AODC	0.77 (0.64)	0.75 (0.69)	0.01 (0.08)	0.13 (0.18)	-0.02 (0.03)	
0.2 ≤ NDVI < 0.4	11 667 461 (3596)						
Land	GRASP/Models AODC	0.76 (0.60)	0.69 (0.69)	0.04 (0.06)	0.13 (0.13)	0.01 (0.03)	
0.4≤NDVI<0.6	7 879 243 (2641)						
Land	GRASP/Models AODC	0.77 (0.65)	0.66 (0.70)	0.04 (0.03)	0.14 (0.08)	-0.01 (0.01)	
NDVI≥0.6	2 766 521 (258)						



Figure 1: Schematic diagram for satellite data selection over (left) land and (right) ocean.



Figure 2: Evaluation of three archives PARASOL/GRASP AOD at 550 nm against AERONET, (a) GRASP/Optimized; (b) GRASP/HP; (c) GRASP/Models. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line. The gray envelope indicates GCOS requirement: max (0.04 or 0.1AOD). The probability density functions of differences (POLDER-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AOD conditions: any AOD, AOD<0.2, 75 0.2≤AOD≤0.7 and AOD>0.7 respectively.



Figure 3: Evaluation of all archive PARASOL/GRASP AE (440/870) against AERONET, (a) GRASP/Optimized; (b) GRASP/HP; (c) GRASP/Models. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line. The probability density functions of differences (POLDER-AERONET) are present in the lower panel.



Figure 4: Evaluation of all archive PARASOL/GRASP AODF at 550 nm with AERONET SDA AODF, (a) GRASP/Optimized; (b) GRASP/HP; (c) GRASP/Models. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line. The gray envelope indicates GCOS requirement applied for AODF: max (0.04 or 0.1AODF). The probability density functions of differences (POLDER-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AODF conditions: any 85 AODF, AODF<0.2, 0.2≤AODF≤0.7 and AODF>0.7 respectively.



Figure 5: The same as Figure 4, but for AODC at 550 nm.



GRASP/HP; (c) GRASP/Models. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line.

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95 Figure 7: Validation of PARASOL/GRASP AOD at 550 nm over land in 2008, (a) GRASP/Optimized; (b) GRASP/HP; (c) GRASP/Models. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line. The gray envelope indicates GCOS requirement: max (0.04 or 0.1AOD). The probability density functions of differences (POLDER-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AOD conditions: any AOD, AOD<0.2, 0.2≤AOD≤0.7 and AOD>0.7 respectively.



Figure 8: Validation of MODIS AOD at 550 nm over land in 2008, (a) DT; (b) DB; (c) MAIAC\_0.1; (d) MAIAC\_0.01. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line. The gray envelope indicates GCOS requirement: max (0.04 or 0.1AOD). The probability density functions of differences (MODIS-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AOD conditions: any AOD, AOD<0.2, 0.2≤AOD≤0.7 and AOD>0.7 respectively.



Figure 9: Validation of PARASOL/GRASP, PARASOL/Operational and MODIS/DT AOD at 550 nm over ocean in 2008. (a) GRASP/Optimized; (b) GRASP/HP; (c) GRASP/Models; (d) Operational; (e) DT. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line. The gray envelope indicates GCOS requirement: max (0.04 or 0.1AOD). The probability
10 density functions of differences (Satellite-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AOD conditions: any AOD, AOD<0.2, 0.2≤AOD≤0.7 and AOD>0.7 respectively.



Figure 10: Spatial distribution of annual mean NDVI for 2008 from GRASP/Models L3 products.



15 Figure 11: Validation of PARASOL/GRASP (a. GRASP/Optimized, b. GRASP/HP and c. GRASP/Models) and MODIS (d. DT and e. DB) AE over land in 2008. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line. The probability density functions of differences (Satellite-AERONET) are present in the lower panel.



Figure 12: Validation of PARASOL/GRASP (a. GRASP/Optimized; b. GRASP/HP and c. GRASP/Models), (d) PARASOL/Operational and (e) MODIS/DT AE over ocean in 2008. The gray dashed line and the red solid line are the 1:1 reference line and the linear regression line. The probability density functions of differences (Satellite-AERONET) are present in the lower panel.



Figure 13: Validation of PARASOL/GRASP (a. GRASP/Optimized; b. GRASP/HP and c. GRASP/Models) and (d) PARASOL/Operational fine mode AOD at 550 nm over land in 2008. The gray envelope indicates GCOS requirement applied for AODF: max (0.04 or 0.1AODF). The probability density functions of differences (POLDER-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AODF conditions: any AODF, AODF<0.2, 0.2≤AODF≤0.7 and AODF>0.7
30 respectively.



Figure 14: Validation of PARASOL/GRASP (a. GRASP/Optimized; b. GRASP/HP and c. GRASP/Models), (d) PARASOL/Operational and (e) MODIS/DT fine mode AOD at 550 nm over ocean in 2008. The gray envelope indicates GCOS requirement applied for AODF:
35 max (0.04 or 0.1AODF). The probability density functions of differences (Satellite-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AODF conditions: any AODF, AODF<0.2, 0.2≤AODF≤0.7 and AODF>0.7 respectively.

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Figure 15: Validation of PARASOL/GRASP (a. GRASP/Optimized; b. GRASP/HP and c. GRASP/Models) coarse mode AOD at 550 nm 40 over land in 2008. The gray envelope indicates GCOS requirement applied for AODC: max (0.04 or 0.1AODC). The probability density functions of differences (POLDER-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AODC conditions: any AODC, AODC<0.2, 0.2≤AODC≤0.7 and AODC>0.7 respectively.



45 Figure 16: Validation of PARASOL/GRASP (a. GRASP/Optimized; b. GRASP/HP and c. GRASP/Models), (d) PARASOL/Operational and (e) MODIS/DT coarse mode AOD at 550 nm over ocean in 2008. The gray envelope indicates GCOS requirement applied for AODC: max (0.04 or 0.1AODC). The probability density functions of differences (POLDER-AERONET) are present in the lower panel. The black, red, blue and green solid lines indicate all AODC conditions: any AODC, AODC<0.2, 0.2≤AODC≤0.7 and AODC>0.7 respectively.



Figure 17: Percentage of the AERONET sites where each product shows the best statistical metric (R, RMSE, BIAS and GCOS Fraction) between 7 PARASOL/GRASP and MODIS AOD products. The number on top of each product is the number of sites where this product has sufficient matchup 2155 points for the comparison.





Figure 18: Maps showing statistical metrics (a) R; (b) RMSE; (c) BIAS; (d) GCOS Fraction (%) for the best performed AOD products (1<sup>st</sup> ranking statistics among 7 PARASOL/GRASP and MODIS products) over each AERONET site. Note that only the 1<sup>st</sup> ranking statistics over each site are 2160 present in the maps.



Figure 19: Percentage of the AERONET sites where each product shows the best statistical metric (R and RMSE) between 5 PARASOL/GRASP and MODIS AE products. The number on top of each product is the number of sites where this product has sufficient matchup points for the comparison.



Figure 20: Maps showing statistical metrics (a) R; (b) RMSE; for the best performed AE products (1<sup>st</sup> ranking statistics among 5 PARASOL/GRASP and MODIS products) over each AERONET site. Note that only the 1<sup>st</sup> ranking statistics over each site are present in the maps.



Figure 21: Spatial distribution of 0.1° x 0.1° seasonal AOD (550 nm) from PARASOL (GRASP/HP and GRASP/Models) and MODIS (DT, DB, and MAIAC) products. DJF – December / January / February; MAM – March / April / May; JJA – June / July / August; SON – September / October / November.



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75 Figure 22: Spatial distribution of 0.1° x 0.1° seasonal AOD (550 nm) differences between PARASOL and MODIS aerosol products, referenced to GRASP/Models.



Figure 23: Spatial distribution of 0.1° x 0.1° seasonal AE from PARASOL (GRASP/HP and GRASP/Models) and MODIS (DT and DB) 80 products.



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85 Figure 25: Spatial distribution of 0.1° x 0.1° seasonal AODF (550 nm) from PARASOL (GRASP/HP and GRASP/Models) and MODIS (DT) products.





Figure 26: Spatial distribution of seasonal AODF (550 nm) differences between PARASOL and MODIS aerosol products, referenced to GRASP/HP.





Figure 27: The same as Figure 25, but for AODC (550 nm).



60°E 120°

60°E

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