Thanks for your helpful comments, we have revised the paper based on your comments. The following is a one-to-one response to your comments.

GENERAL COMMENTS

This manuscript by Zhang et al. conducted the fine mode fraction (FMF) retrieval from muliti-angular polarimeter (PARASOL). Technically, the total AOD is determined from intensity measurements, and fine mode AOD is derived from multi-angular polarized measurements. Then the ratio of AOD and fine mode AOD derives FMF. This method generally sounds, and has been published in Zhang et al. (2017, 2018). This manuscript is mainly focus on the validation of retrieved FMF using AERONET, MODIS, PARASOL/GRASP products. The main concern here is that each product may have different definition of their FMF, this should be fully considered before conducting validation and inter-comparison. For example, MODIS FMF over land is the ratio to reflectance instead of total AOD; therefore MODIS FMF over land has little physical meaning. Over ocean, by single scattering approximation, FMF can be approximated as weighted for AOD (see discussions in Remer et al., 2005). Additionally, the objective is not clear why the authors pay close attention to FMF instead of fine mode AOD, the uncertainties in both AOD and fine AOD could significantly worsen the FMF quality, and a good FMF doesn't necessarily produce a good estimation of fine mode AOD. Overall, I think this manuscript is within the scope of AMT. Some comments and concerns are required to be addressed and clearly stated before being published. The specific comments are listed as follow.

In the revised paper, we have discussed the differences in the definition of different FMF products. Please check our revised paper later. This part is also included in our answer to your comment below. In 2015, we proposed the PMRS model (Zhang et al., 2015), which is a model based on physical methods to estimate PM2.5 concentration. In that model, FMF is an important input parameter and cannot be replaced by AOD_f. Since the existing MODIS FMF products are difficult to meet the application requirements of the PMRS model, we started the research of using multi-angle polarization sensors to retrieve FMF. In addition, FMF can also be used to distinguish anthropogenic and natural aerosol types (Bellouin et al., 2005). We think that FMF is also important for research in the field of atmospheric environment.

References:

Zhang, Y., and Li, Z.: Remote sensing of atmospheric fine particulate matter (PM2.5) mass concentration near the ground from satellite observation, Remote Sensing of Environment, 160, 252-262, 10.1016/j.rse.2015.02.005, 2015.

Bellouin, N., Boucher, O., Haywood, J., Reddy, M.S., 2005. Global estimates of aerosol direct radiative forcing from satellite measurements. Nature 438, 1138–1141.

SPECIFIC COMMENTS

Line 39: please be cautious to interpret MODIS FMF over land, it is weighted of reflectance instead of AOD (see discussions in Remer et al., 2005; Chen et al., 2020);

Answer: After we read the comments of you and another reviewer, we realized that we had a misunderstanding of MODIS FMF. We have rewritten this paragraph as follows:

However, other new aerosol optical parameters, such as the fine-mode fraction (FMF), are quite different in definition from the ground-based observations (Remer et al., 2005;Levy et al., 2010), which makes them incomparable.

References:
Remer, L. A., Kaufman, Y. J., Tanré, D., Mattoo, S., Chu, D. A., Martins, J. V., Li, R. R., Ichoku, C., Levy, R. C., and Kleidman, R. G.: The MODIS Aerosol Algorithm, Products, and Validation, Journal of the Atmospheric Sciences, 62, 947-973, 2005.
Levy, R. C., Remer, L. A., Kleidman, R. G., and Mattoo, S.: Global evaluation of the Collection 5 MODIS dark-target aerosol products over land, Atmospheric Chemistry & Physics, 10, 10399-10420, 2010.

Line 53: This is not true. Please check Chen et al., 2020 (10.5194/essd-2020-224).

Answer: Our expression was not clear. We wanted to say that LOA only provides AOD_f in its operational aerosol products over land. Chen et al. also mentioned this information in their section 4.1 (10.5194/essd-2020-224). We have rewritten this paragraph as follows:

For example, the French Laboratoire d'Optique Atmospherique (LOA) only provided the fine-mode aerosol optical depth (AODf) datasets in its operational product over land (Deuzé et al., 2001;Tanré et al., 2011), the total aerosol optical depth (AODt) was not provided (Chen et al., 2020).

References:
Deuzé, J. L., Bréon, F. M., Devaux, C., Goloub, P., Herman, M., Lafrance, B., Maignan, F., Marchand, A., Nadal, F., Perry, G., and Tanré, D.: Remote sensing of aerosols over land surfaces from POLDER-ADEOS-1 polarized measurements, Journal of Geophysical Research, 106, 4913, 10.1029/2000jd900364, 2001.
Tanré, D., Bréon, F. M., Deuzé, J. L., Dubovik, O., Ducos, F., François, P., Goloub, P., Herman, M., Lifermann, A., and Waquet, F.: Remote sensing of aerosols by using polarized, directional and spectral measurements within the A-Train: the PARASOL mission, Atmospheric Measurement Techniques, 4, 1383-1395, 10.5194/amt-4-1383-2011, 2011.
Chen, C., Dubovik, O., Fuertes, D., Litvinov, P., Lapyonok, T., Lopatin, A., Ducos, F., Derimian, Y., Herman, M., Tanré, D., Remer, L. A., Lyapustin, A., Sayer, A. M., Levy, R. C., Hsu, N. C., Descloitres, J., Li, L., Torres, B., Karol, Y., Herrera, M., Herreras, M., Aspetsberger, M., Wanzenboeck, M., Bindreiter, L., Marth, D., Hangler, A., and Federspiel, C.: Validation of GRASP algorithm product from POLDER/PARASOL data and assessment of multi-angular polarimetry potential for aerosol monitoring, Earth Syst. Sci. Data Discuss., 2020, 1-108, 10.5194/essd-2020-224, 2020.

Line 71-72: 'there is a problem of low retrieval value for high aerosol loading' ??? Could you specify it, underestimation for high AOD or FMF?

Answer: The underestimation is for AOD_f for high aerosol loading. We have rewritten this sentence as follows:

In polarization retrieval, the problem of a low AOD_f retrieval value for high aerosol loading exists

Line 82: thesis?? -> study.

Answer: We have corrected it.

Line 148: 3x3 window ? is it equivalent to 3x18km?

Answer: Yes, it is equivalent to 3x18km, which is about 54 km. We have added this information as follows:

*The satellite retrieval result used for comparison is the effective retrieval result centred on the location of the AERONET site within the closest distance in the 3*3 window (about 54 km).*

Line 154: is there any intention or reference to use ±0.1±10% EE for FMF?

Answer: The other reviewer also mentioned this issue. However, there does not seem to be a unified standard for EE definition of FMF, different studies have different standards. For example, the study of Cheng et al. did not define the EE of FMF. The study of Yan et al. defined the EE of FMF as ±0.4. The study of Chen et al. defines three types of FMF EE: +/-(0 +40%), +/-(0 +25%), +/-(0.03 +20%). We have reconsidered the definition of EE for FMF. Firstly, we believe that the EE of FMF should not increase as the value increases, which is different from AOD. Secondly, the ground-based FMF has a certain error. According to the research of O'Neill et al., the SDA method has an uncertainty of about 0.1. We considered the absolute error part (0.1) of the previous EE of FMF and the uncertainty (0.1) of the ground-based FMF, and finally changed the EE of FMF in this study to ±0.2.

References:

T, Cheng, X, et al. Aerosol optical depth and fine-mode fraction retrieval over East Asia using multi-angular total and polarized remote sensing[J]. Atmospheric Measurement Techniques, 2012.

Yan X, Li Z , Shi W , et al. An improved algorithm for retrieving the fine-mode fraction of aerosol optical thickness, part 1: Algorithm development[J]. Remote Sensing of Environment, 2017, 192:87-97.

Chen, X., de Leeuw, G., Arola, A., Liu, S., Liu, Y., Li, Z., and Zhang, K.: Joint retrieval of the aerosol fine mode fraction and optical depth using MODIS spectral reflectance over northern and eastern China: Artificial neural network method, Remote Sensing of Environment, 249, 112006, 2020

O'Neill, Norm T, Dubovik, et al. Modified Ångström Exponent for the Characterization of Submicrometer Aerosols[J]. Applied Optics, 2001.

Line 158: Section name is wrong.

Answer: We have modified the section name as 'Validation against AERONET ground-based data'.

Figure 3: is this all points from 2006-2013? Any filter scheme used, please clarify.

Answer: Yes, this is all the matched points from 2006 to 2013. When the retrieved AOD_f is greater than the retrieved AOD_t, we consider this situation as a failure of the FMF retrieval, and the results of this part were not involved in the comparison. These results account for about 10% We have added those information in section 2.3 as follows:

Note that when the retrieved AOD_f is greater than the retrieved AOD_t, we consider this situation as a failure of the FMF retrieval, and the results of this part were not involved in the comparison. These results account for about 10%.

Line 177: errors : : : are stable: : : ?? please consider 'uncertainty'.

Answer: We have corrected it.

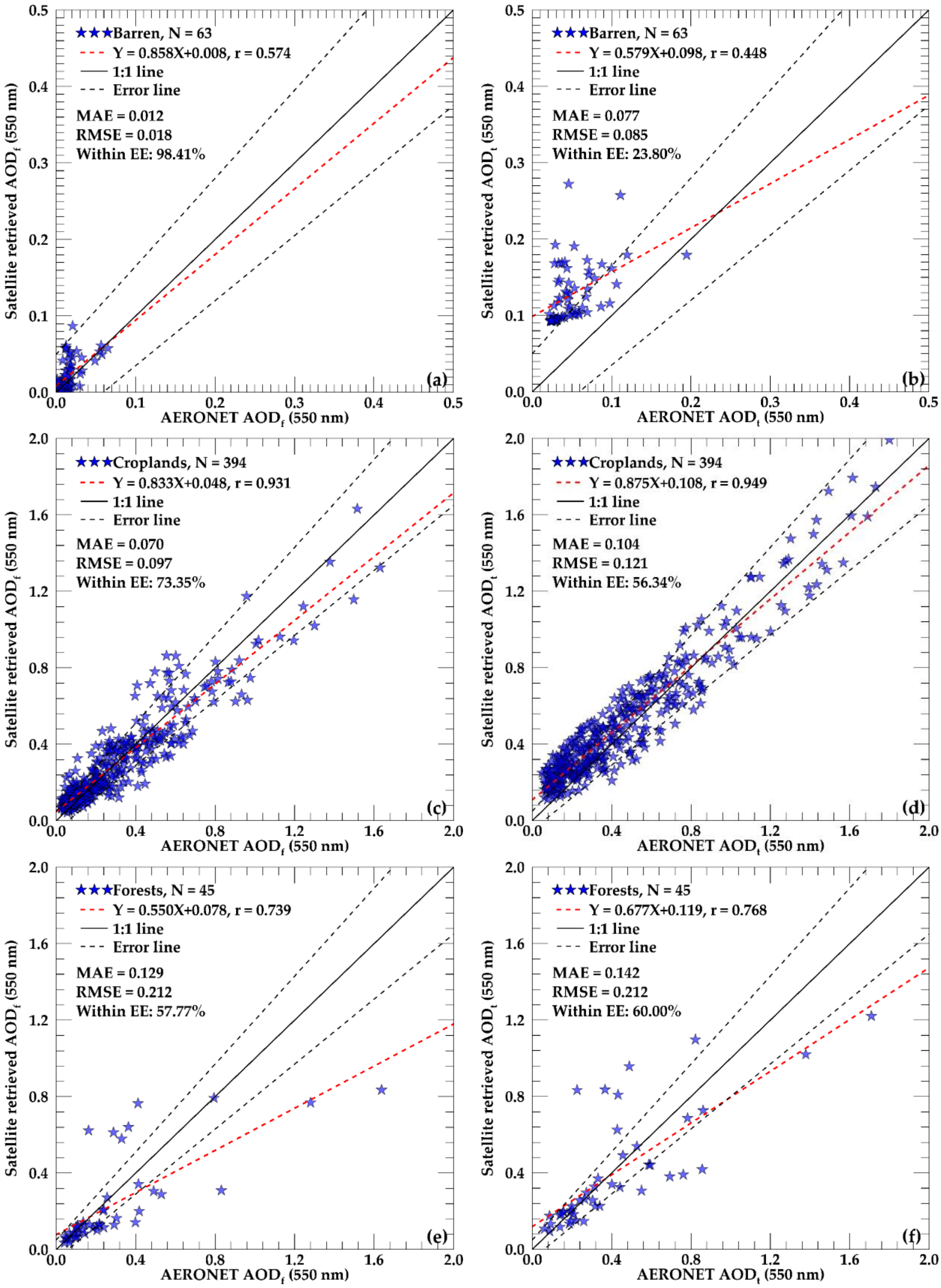
Line 182: the definitions of AERONET FMF and retrieved AODf/AOD are not identical.

Answer: We agree that the definitions of the AERONET FMF and retrieved FMF are not identical. However, they have some similarities. The definition of the AOD_f in our study is indefinite, and it has no clear cut-off particle size. Similarly, there is also no clear definition of AOD_f in the ground-based SDA algorithm. Therefore, we think that although they are not equivalent, the two are comparable. We prefer to use the SDA FMF as the 'truth value' for validation.

Table 3: Number of points is critical, as well as other parameters (r, rmse, etc.).

Answer: We have added the information of the number of points, r and bias. According to the comments from the other reviewer, we also added the comparison between the AOD_f and AOD_t retrieval results and ground-based observations of different surface types. The relevant contents are shown as below:

Since our FMF is obtained from the ratio of AOD_f and AOD_t retrieval results, and the retrieval accuracy of the two parameters directly determines the retrieval accuracy of FMF, we further compared the retrieved AODs at the six different surface types with those of the ground-based data from 2006 to 2013, and the statistical results are shown in Figure R1 and Table R1. It can be seen from Figure R1 that for the comparison results of AOD_f, except for the barren type, the AOD_f at all surface types are in good agreement with the ground-based observation results, and the r is greater than 0.7. Because the data of the barren type mainly come from the QOMS_CAS site, the AOD_f value at this site is low, and the r is not suitable for evaluating the retrieval performance. Most of the retrieval results at barren type fall within the EE, which can indicate that the retrieval results at this type have a good accuracy. For the comparison results of AOD_t, the retrieval results at barren type are obviously positively shifted. This is due to the low aerosol loading at the QOMS_CAS site, and the inaccurate estimation of the surface reflectance can easily magnify the errors in the retrieval results. It indicates that the EOF method used to retrieve AOD_t in this study still needs further improvement. However, it is difficult to analyse the reasons for the negative bias of most FMF retrieval results from the scatter plot, so we further counted the biases of AOD_t and AOD_f. Table R1 shows that the bias of the retrieved AOD_f and AOD_t at the six different surface types. It can be seen from Table R1 that the proportion of positive bias is greater than the proportion of negative offset for most AOD_t retrieval results, while AOD_f is the opposite. For the overall result, the bias of AOD_f is -0.037, where the proportion of negative bias is 58.68%, and the bias of AOD_t is 0.063, where the proportion of positive bias is 68.29%, indicating that the AOD_f retrieval result has a negative bias, and the AOD_t retrieval result has a positive bias, that is, the numerator is small and the denominator is large, eventually leading to a negative bias of FMF.


Barren, N = 63
Y = 0.858X+0.008, r = 0.574
1:1 line
Error line
MAE = 0.012
RMSE = 0.018
Within EE: 98.41%
(a)
Satellite retrieved AOD$_f$ (550 nm)
AERONET AOD$_f$ (550 nm)
Barren, N = 63
Y = 0.579X+0.098, r = 0.448
1:1 line
Error line
MAE = 0.077
RMSE = 0.085
Within EE: 23.80%
(b)
Satellite retrieved AOD$_t$ (550 nm)
AERONET AOD$_t$ (550 nm)
Croplands, N = 394
Y = 0.833X+0.048, r = 0.931
1:1 line
Error line
MAE = 0.070
RMSE = 0.097
Within EE: 73.35%
(c)
Satellite retrieved AOD$_f$ (550 nm)
AERONET AOD$_f$ (550 nm)
Croplands, N = 394
Y = 0.875X+0.108, r = 0.949
1:1 line
Error line
MAE = 0.104
RMSE = 0.121
Within EE: 56.34%
(d)
Satellite retrieved AOD$_t$ (550 nm)
AERONET AOD$_t$ (550 nm)
Forests, N = 45
Y = 0.550X+0.078, r = 0.739
1:1 line
Error line
MAE = 0.129
RMSE = 0.212
Within EE: 57.77%
(e)
Satellite retrieved AOD$_f$ (550 nm)
AERONET AOD$_f$ (550 nm)
Forests, N = 45
Y = 0.677X+0.119, r = 0.768
1:1 line
Error line
MAE = 0.142
RMSE = 0.212
Within EE: 60.00%
(f)
Satellite retrieved AOD$_t$ (550 nm)
AERONET AOD$_t$ (550 nm)

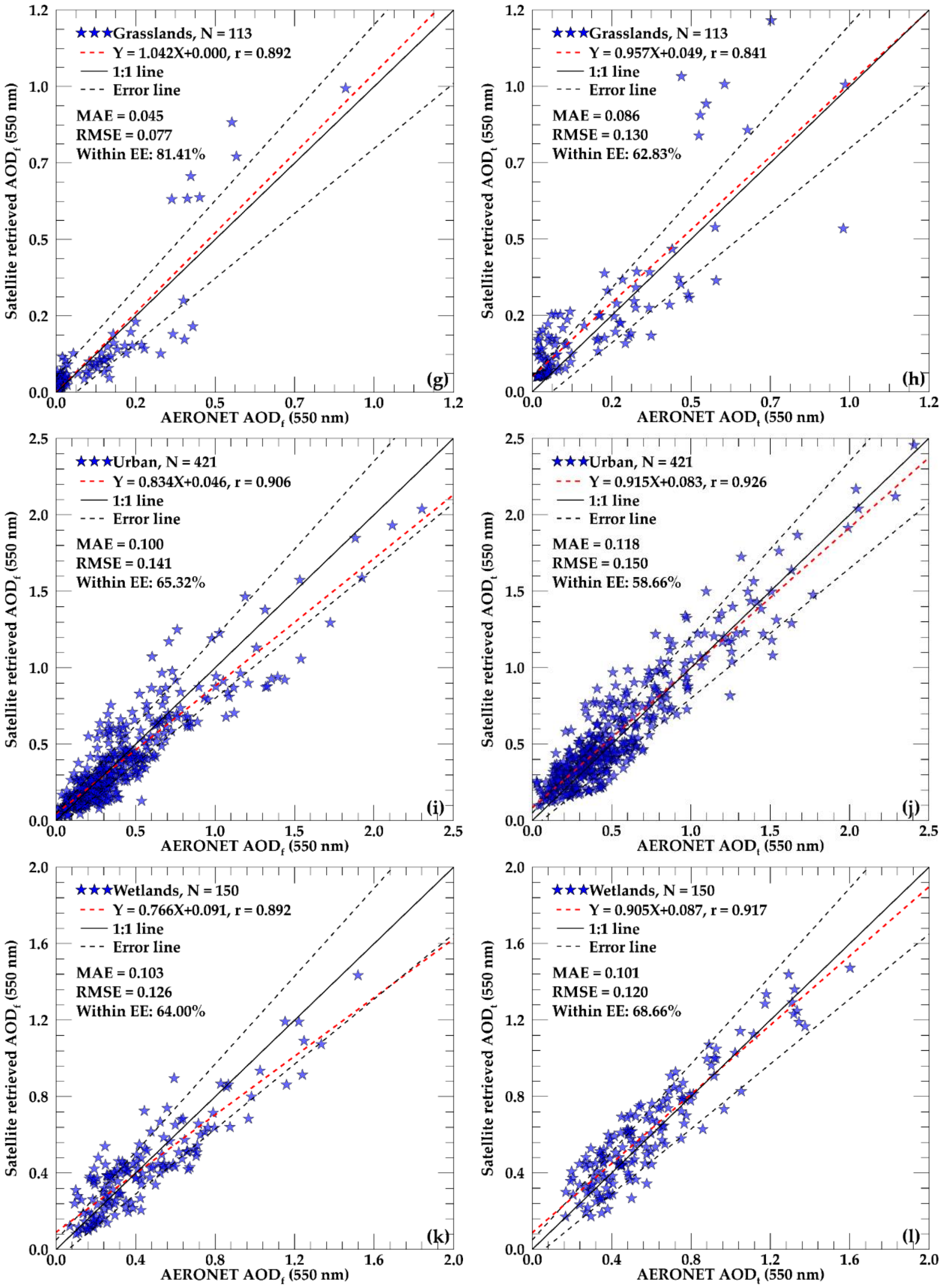


Figure R1. AODs results comparison of 6 surface types. (a), (c), (e), (g), (i), and (k) are the AOD$_t$ validation results for the type of barren, croplands, forests, grasslands, urban, and wetlands, respectively. (b), (d), (f), (h), (j), and (l) are the AOD$_f$ validation results for the type of barren, croplands, forests, grasslands, urban, and wetlands, respectively.

The final revised table is shown as below:

Table R1. Statistical analysis of AOD$_f$ and AOD$_t$ bias

Land cover	Retrieval	N	r	Bias	Proportion of	Proportion of

type	parameter (550 nm)				negative bias	positive bias
Barren	AOD_f	63	0.574	0.006	44.44%	55.56%
	AOD_t		0.448	0.111	1.59%	98.41%
	FMF		0.711	-0.144	87.30%	12.70%
Croplands	AOD_f	394	0.931	-0.038	55.84%	44.16%
	AOD_t		0.949	0.077	27.16%	72.84%
	FMF		0.651	-0.064	64.47%	35.53%
Forests	AOD_f	45	0.739	-0.049	64.44%	35.56%
	AOD_t		0.768	-0.019	48.89%	51.11%
	FMF		0.831	-0.102	75.56%	24.44%
Grasslands	AOD_f	113	0.892	0.007	38.05%	61.95%
	AOD_t		0.841	0.061	23.89%	76.11%
	FMF		0.777	-0.033	55.75%	44.25%
Urban	AOD_f	421	0.906	-0.043	64.61%	35.39%
	AOD_t		0.926	0.057	38.72%	61.28%
	FMF		0.733	-0.079	72.45%	27.55%
Wetlands	AOD_f	150	0.892	-0.065	69.33%	30.67%
	AOD_t		0.917	0.048	37.33%	62.67%
	FMF		0.508	-0.031	55.33%	44.67%
Overall	AOD_f	1186	0.868	-0.037	58.68%	41.32%
	AOD_t		0.867	0.063	31.71%	68.29%
	FMF		0.770	-0.068	66.95%	33.05%

Line 220: Please identify products name and version, and last access, etc. (This is necessary for all products used in the manuscript)

Answer: We have added the GRASP products information as follows:

The GRASP product version we processed is V2.06, which is the latest version that can be obtained from AERIS/ICARE Data and Services Center (http://www.icare.univ-lille.fr, last accessed on December 27, 2020).

Line 222: what do you mean normalized FMF?

Answer: To facilitate the comparison of the differences in the spatial distribution trends of those results from this study, MODIS and GRASP, all the results are normalized, meaning they are divided by the maximum value in the respective FMF image.

Section 3.3: why only 2013 data is compared? It would be interesting to check more data 2006-2013 and other related parameters, e.g. AOD and fine mode AOD, to make the conclusion more solid.

Answer: Due to the limited ground PM2.5/PM10 data, we can only compare the results in 2013. As shown in Figure R1 and Table R1, we compared the retrieved AOD_f and AOD_t with those from the ground-based observations. We also added the following discussion about FMF definitions of this study and GRASP in Section 3.3:

GRASP products provide AOD_f and AOD_t datasets, but do not directly provide FMF datasets. In this study, the ratio of the two was used to obtain the GRASP FMF. However, it should be *noted that the definition of GRASP AOD_f is somewhat different from the AOD_f in our research, which may eventually lead to the difference in the definition of FMF. The AOD_f in our study is similar to the definition in the ground-based SDA algorithm; there is no clear cut-off particle size, that is, its definition is indefinite. This is different from the AOD_f obtained by calculating and integrating the size distribution in GRASP, so the difference in the spatial distribution results of the two may be caused by the definition, rather than a problem in the retrieval algorithm. In the research of Chen et al. (Chen et al., 2020), in their comparison with AERONET observations, the r of AOD_f is between 0.868 (models approach) and 0.924 (high-precision approach), which is similar to the r (0.868) of AOD_f in this study, but their bias is only -0.02 (models approach) and 0.01 (high-precision approach), which is different from the bias (-0.037) of AOD_f in this study. This indicates that the definition of AOD_f in GRASP and our study may be different.*

Reference:

Chen, C., Dubovik, O., Fuertes, D., Litvinov, P., Lapyonok, T., Lopatin, A., Ducos, F., Derimian, Y., Herman, M., Tanré, D., Remer, L. A., Lyapustin, A., Sayer, A. M., Levy, R. C., Hsu, N. C., Descloitres, J., Li, L., Torres, B., Karol, Y., Herrera, M., Herreras, M., Aspetsberger, M., Wanzenboeck, M., Bindreiter, L., Marth, D., Hangler, A., and Federspiel, C.: Validation of GRASP algorithm product from POLDER/PARASOL data and assessment of multi-angular polarimetry potential for aerosol monitoring, Earth Syst. Sci. Data Discuss., 2020, 1-108, 10.5194/essd-2020-224, 2020.

Figures 6, 7, 8: it is important to mention the spatial resolution, visually, the derived FMF in figure 6 has much coarser resolution than others.

Answer: We have added the spatial resolution information of the corresponding result in the figure title. According to the suggestion from the other reviewer, we integrated the original Figure 6-9 into one Figure (Figure R2).

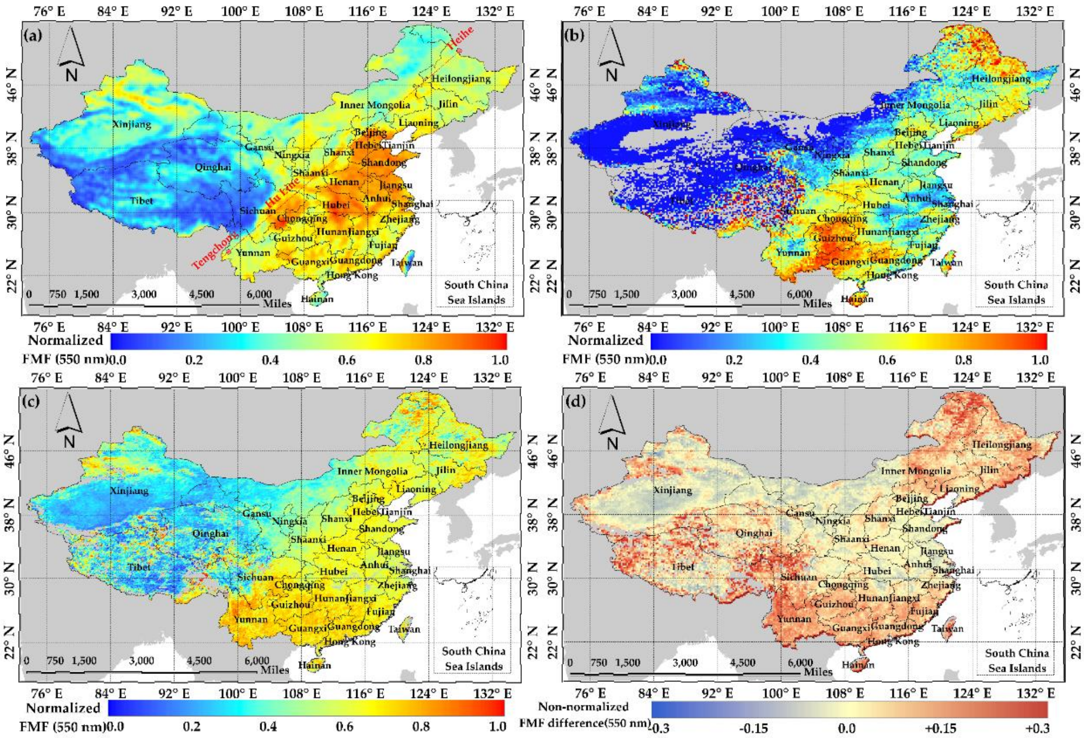


Figure R2. Distribution of FMF of China in 2013 from different sources. (a) is the normalized results of this study (18 km resolution), (b) is the normalized results of MODIS (10 km resolution), (c) is the normalized results of GRASP (6 km resolution), and (d) is the GRASP results minus the retrieved results (non-normalized, 18 km resolution).

Figures9, 16: the quality of figures showing differences can be improved by using more adequate colorbar.

Answer: There are 5-6 points labeled on the color scale now (Figure R2). However, we only retained the seasonal average spatial distribution results of FMF in the revised paper according to the comments from the other reviewer, and the original Figure 16 has been deleted.

Line 346: throughout the manuscript, no place specified the MODIS (TERRA or AQUA or both) dataset.

Answer: We have added the information of the MODIS FMF results in section 3.2 as follows:

The MODIS FMF results were derived from the MYD04 product of collection 6.1.

Line 370: Is there any specific reason to pay close attention to FMF instead of fine mode AOD? On one hand, the uncertainties in both AOD and fine AOD could significantly worsen the FMF, on the other hand, a good FMF doesn't necessarily produce a good estimation of fine mode AOD, which can compensate by AOD and fine AOD, right?

Answer: In 2015, we proposed the PMRS model (Zhang et al., 2015), which is a model based on physical methods to estimate $PM_{2.5}$ concentration. In that model, FMF is an important input parameter and cannot be replaced by AOD_f. Since the existing MODIS FMF products are difficult to meet the application requirements of the PMRS model, we started the research of using multi-angle polarization sensors to retrieve FMF. In addition, FMF can also be used to distinguish anthropogenic and natural aerosol types (Bellouin et al., 2005). We think that FMF is also important for research in the field of atmospheric environment. We have rewritten that sentence as follows: *In the future, it is still necessary to further improve the retrieval accuracy of AODf and AODt. to obtain more accurate FMF results. In this way, some applications that rely on FMF (such as using the PMRS model to estimate PM2.5 concentration) can have better performance.*

References:

Zhang, Y., and Li, Z.: Remote sensing of atmospheric fine particulate matter (PM2.5) mass concentration near the ground from satellite observation, Remote Sensing of Environment, 160, 252-262, 10.1016/j.rse.2015.02.005, 2015.

Bellouin, N., Boucher, O., Haywood, J., Reddy, M.S., 2005. Global estimates of aerosol direct radiative forcing from satellite measurements. Nature 438, 1138–1141.