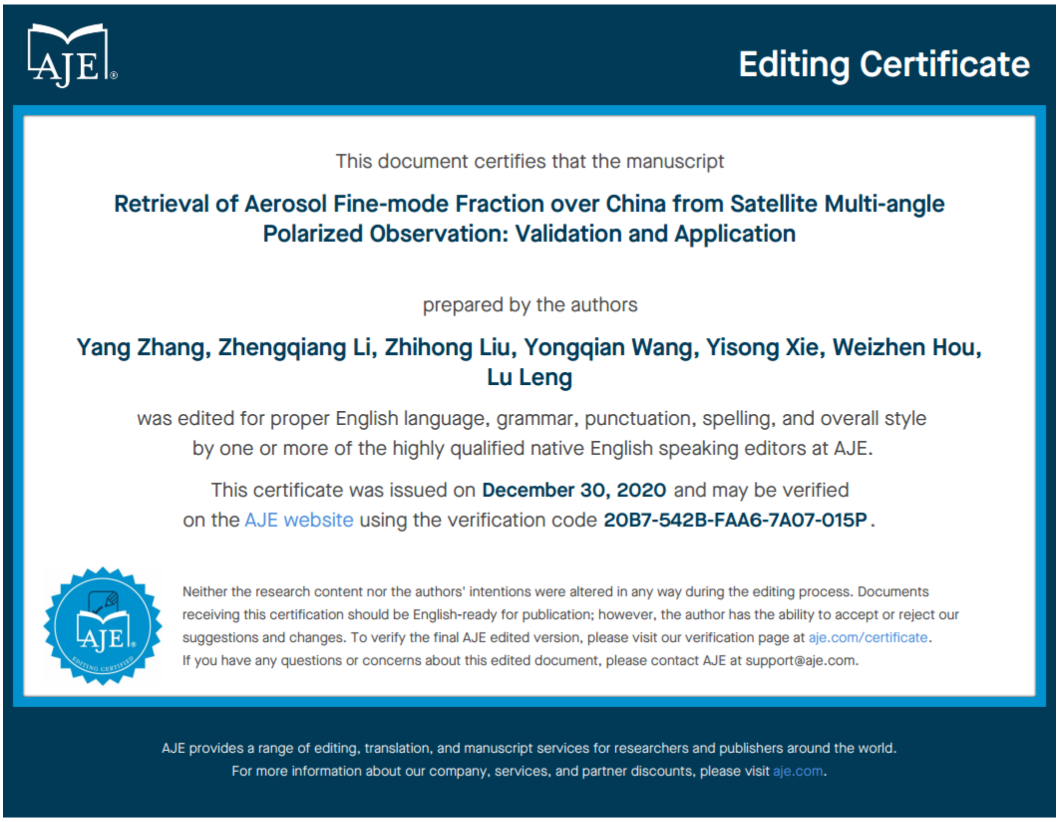
Responses to reviewer 1

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.

Comment: This is an interesting study looking at the fine mode fraction (FMF) of aerosol optical depth in China. The main focus is the comparison between results from a previously published algorithm of the authors applied to POLDER data (referred to in the paper as "the algorithm"), the GRASP retrieval applied to POLDER, the MODIS Dark Target land retrieval, and the AERONET spectral deconvolution algorithm (SDA) and almucantar scan size distribution (SD) retrieval algorithm. The topic is relevant to the journal and the Special Issue. After total AOD, fine vs. coarse AOD is one of the next main frontiers of interest.

The quality of language needs some improvement. I appreciate the authors' first language is not English, and they have done a good job explaining what was done in the analysis. But some copy-editing will be necessary to bring the article to publication standards as phrasing is strange in places (too many to go through as a reviewer). This might be able to be handled by the journal production office, but if the authors have access to a service or colleague who is able to give a proof-read that would be beneficial as well. Again, the authors have done a pretty good job with the writing overall.

Answer: We submitted the manuscript to American Journal Experts (AJE) for editing service before submission, but we made some changes to the manuscript after the editing service was completed. This may be the cause of difficulty in reading some sentences. We have submitted the revised manuscript to AJE for editing again. Thanks for your understanding.



Editing Certificate

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Retrieval of Aerosol Fine-mode Fraction over China from Satellite Multi-angle Polarized Observation: Validation and Application

prepared by the authors

Yang Zhang, Zhengqiang Li, Zhihong Liu, Yongqian Wang, Yisong Xie, Weizhen Hou, Lu Leng

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Figure R1. The editing certificate of AJE

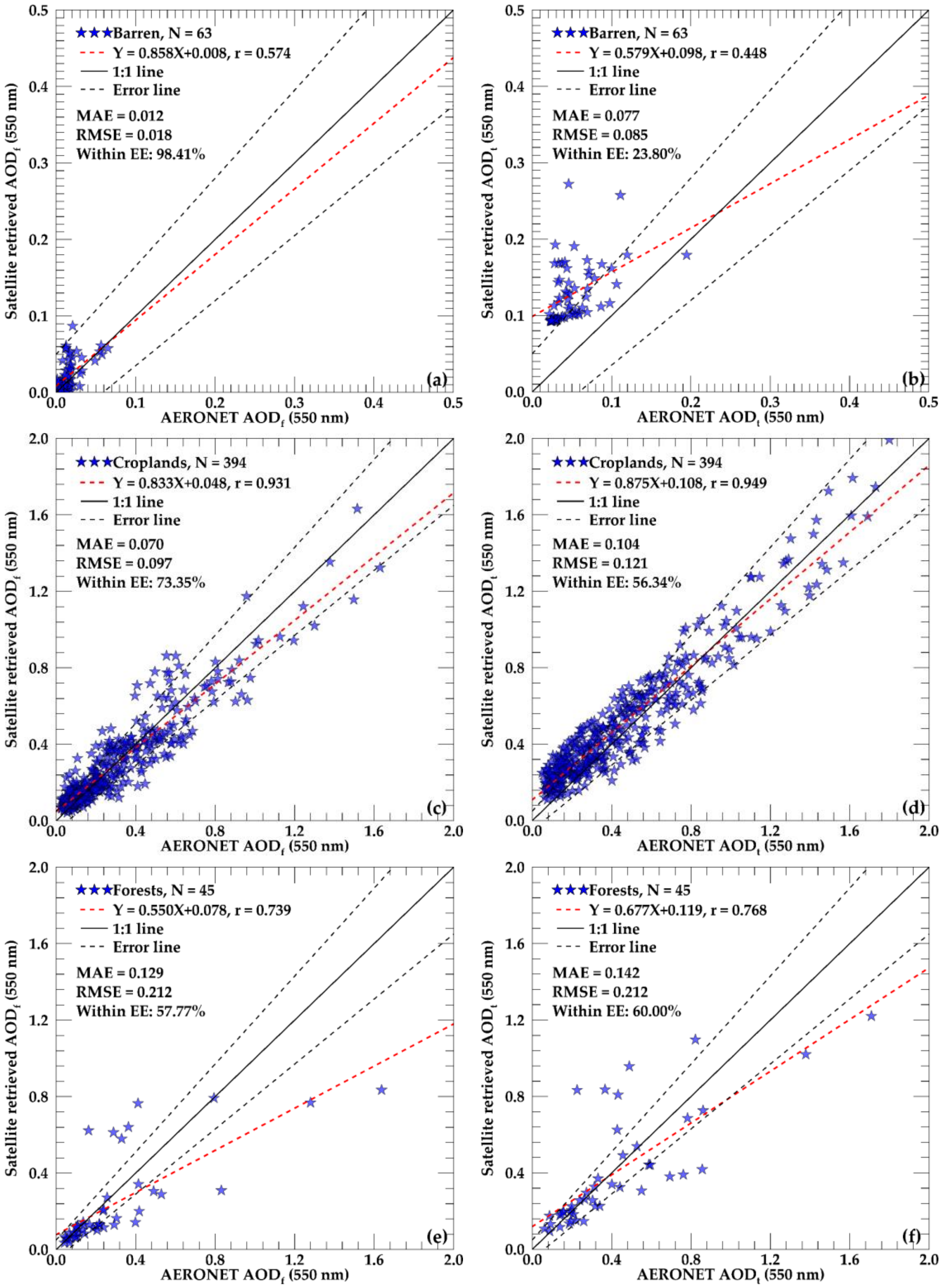
My recommendation is for major revisions, and I would like to review the revised version.

Major comments:

1. It is not just the FMF which is of interest, but the overall fine and coarse AOD. It seems like a missed opportunity not to evaluate e.g. the fine mode and total AOD as well. Looking at only FMF we don't know if a bias in that is because the retrievals have errors in the total AOD or just the ratio between modes. This is briefly shown (Table 3) but only via summary metrics (would be good to see the data points) and only for the authors' approach (not MODIS or GRASP). I recommend the authors add this in the revised version. These could be also split by, for example, aerosol type or surface type (as these are some factors which often affect retrieval performance).

Answer: Thanks for your suggestion. We re-analyzed the corresponding AODf and AODt retrieval results from 2006 to 2013, instead of the previous result in 2013, and added the following content:

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 R2 and Table R1. It can be seen from Figure R2 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%
Satellite retrieved AOD_f (550 nm)
AERONET AOD_f (550 nm)
(a)
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%
Satellite retrieved AOD_t (550 nm)
AERONET AOD_t (550 nm)
(b)
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%
Satellite retrieved AOD_f (550 nm)
AERONET AOD_f (550 nm)
(c)
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%
Satellite retrieved AOD_t (550 nm)
AERONET AOD_t (550 nm)
(d)
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%
Satellite retrieved AOD_f (550 nm)
AERONET AOD_f (550 nm)
(e)
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%
Satellite retrieved AOD_t (550 nm)
AERONET AOD_t (550 nm)
(f)

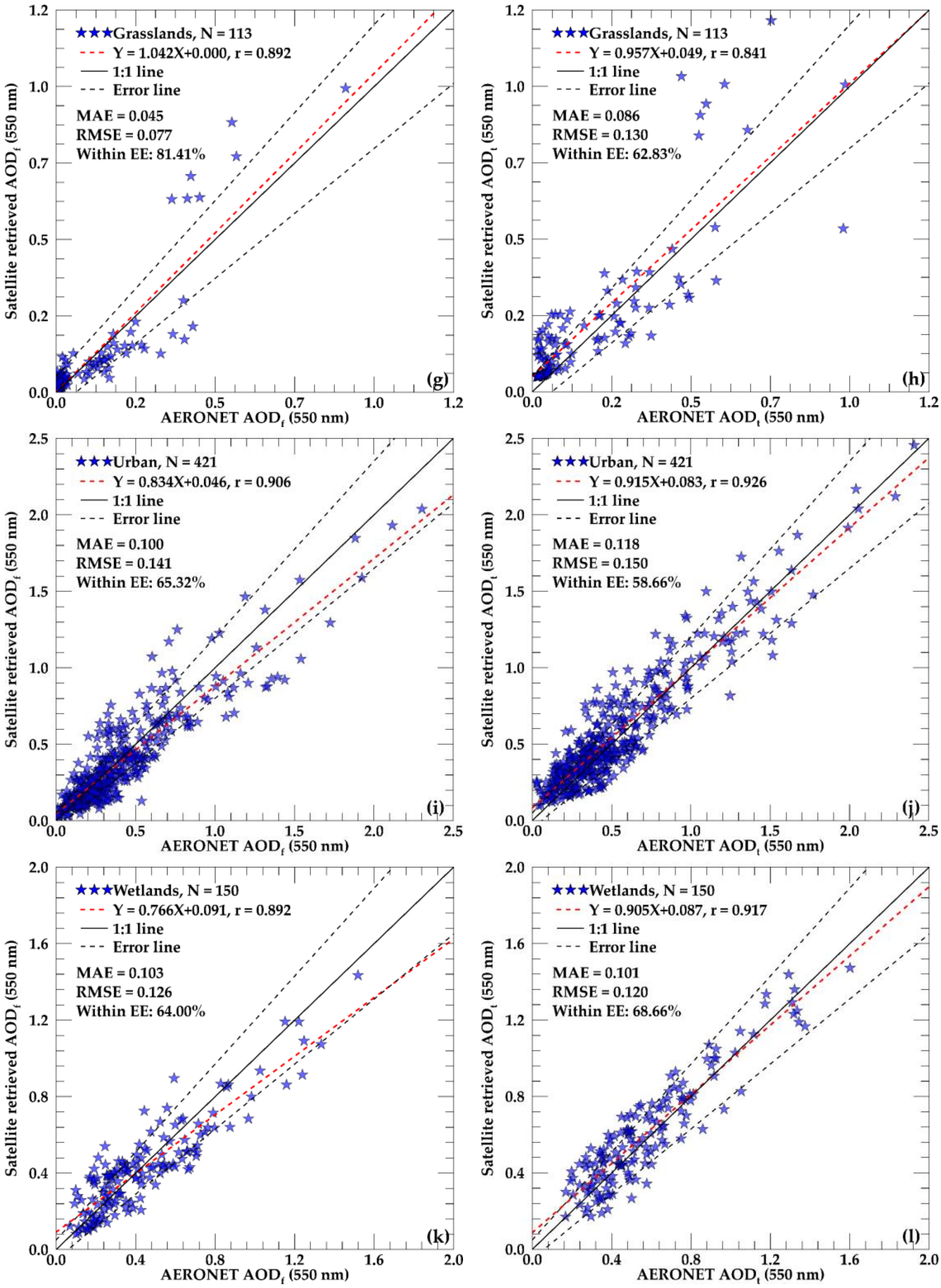


Figure R2. AODs results comparison of 6 surface types. (a), (c), (e), (g), (i), and (k) are the AOD_t validation results for the type of barren, croplands, forests, grasslands, urban, and wetlands, respectively. (b), (d), (f), (h), (j), and (l) are the AOD_f validation results for the type of barren, croplands, forests, grasslands, urban, and wetlands, respectively.

Table R1. Statistical analysis of AOD_f and AOD_t bias

Land cover type	Retrieval parameter (550 nm)	N	r	Bias	Proportion of negative bias	Proportion of 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%

2. The authors' interpretation of MODIS Dark Target land FMF is, to my knowledge, not correct. The MODIS land fine weighting parameter eta is not a "fine mode fraction" but a "fine model fraction". The Dark Target land retrieval mixes between two bimodal size distributions, one which is mostly fine mode and one which is mostly coarse mode. See Levy 2007 (https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2006JD007815), especially Figure 4 there. So in MODIS, FMF=0 does not mean no fine mode aerosol. It means that the proportion of the bimodal fine-dominated optical model is zero. There is still some fine mode aerosol from the coarse-dominated optical model. Similarly FMF=1 in MODIS still has some coarse aerosol present. This misinterpretation affects all the discussion of MODIS results.

Answer: Thanks for your correction, we do have a wrong understanding of the FMF definition of MODIS. We have rewritten the discussion about the two, and the revised part is shown as below:

MODIS aerosol products also include FMF data sets, but this FMF has a different definition. In fact, the FMF of MODIS refers to the 'fine model fraction', which is the proportion of bimodal fine-dominated aerosol model, but not pure fine mode (Levy et al., 2007). Because the FMF results obtained by MODIS are different in definition from the ground-based results (Levy et al., 2009), the retrieval results are quite different from the ground-based observation results, which limits the research that depends on the FMF parameter. We compared the retrieved and

MODIS FMF with the AERONET ground-based observations to further evaluate the significance of our results. The MODIS FMF results were derived from the MYD04 product of collection 6.1. Figure 7 shows the comparison between the two results and the AERONET ground-based observation results from 2011 to 2013, which is the results where both MODIS and POLDER matching the ground-based observations. As seen from the figure, compared with ground-based observations, the r of FMF obtained in this study is 0.812, while that of MODIS is 0.302. The correlation coefficient of the results obtained in this study is much higher than that of MODIS. At the same time, notice that there are many 0 values in the MODIS results. These 0 values are not meaningless but correspond to the situation where there is no the fine-dominated aerosol model.

More statistical results of the two are shown in Table 4. The table shows that the FMF results obtained in this study have an MAE of 0.072, an RMSE of 0.102, and a Within EE of 87.41%, whereas results of MODIS have an MAE of 0.512, RMSE of 0.574, and Within EE of 19.58%. The statistical indicators of the FMF results obtained by our study are closer to the ground-based observations than the MODIS results. Nevertheless, note that this does not mean that the FMF of MODIS has a large deviation. As mentioned above, there is a difference in definition between the FMF of MODIS and the ground-based observations; consequently, it is difficult to obtain the true deviation of MODIS FMF based on ground-based observations.

References:

Levy, R. C., Remer, L. A., and Dubovik, O.: Global aerosol optical properties and application to Moderate Resolution Imaging Spectroradiometer aerosol retrieval over land, Journal of Geophysical Research: Atmospheres, 112, https://doi.org/10.1029/2006JD007815, 2007.

Levy, R. C., Remer, L. A., Tanré, D., Mattoo, S., Vermote, E. F., and Kaufman, Y. J.: Algorithm for Remote Sensing of Tropospheric Aerosol over Dark Targets from MODIS:Collections 005 and 051: Revision 2, 2009.

3. The definition of FMF in other products is not be the same either, e.g. the AERONET SDA assumes a combination of fine and coarse modes but the AERONET sky-scan retrieval and (I think) GRASP look for a minimum in the size distribution and make the fine/coarse split there. The manuscript should be more detailed about the exact definition of fine mode fraction within the products, and make sure they are comparable. If not then the discrepancies will be partly due to definition differences rather than retrieval problems.

Answer: We agree that different products in this study have different FMF definitions, We have added the following discussion about FMF definitions 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.

Minor comments:

1. Line 98: is the EOF method similar to the MISR land approach? That could be mentioned (and compared if different) as it is likely that the readership of this journal would have some familiarity with it.

Answer: Yes. The EOF method used in our retrieval is similar to the MISR approach, we transplanted this method to POLDER. We have added the following relevant information in the revised paper:

The EOF method has previously been used for the retrieval of land aerosols on Multi-angle Imaging Spectro Radiometer (MISR); we transplanted this method to POLDER based on the MISR approach. For more details, please refer to our 2017 study (Zhang et al., 2017)

References:

Zhang, Y., Li, Z., Qie, L., Hou, W., Liu, Z., Zhang, Y., Xie, Y., Chen, X., and Xu, H.: Retrieval of Aerosol Optical Depth Using the Empirical Orthogonal Functions (EOFs) Based on PARASOL Multi-Angle Intensity Data, Remote Sensing, 2017, 578, 2017.

2. Line 133: Angstrom should be written Ångström here.

Answer: We have modified 'Angstrom' to 'Ångström'.

3. Line 154: I don't know why it makes sense for EE to be +/-(0.1+10%). Why should FMF uncertainty depend on FMF? Is high FMF slightly harder to retrieve? More justification is needed here. If this was used in a previous study, we need to see the justification there, and if there wasn't one, then that's an issue. I do not see a physical reasoning why FMF uncertainty should be a function of FMF.

Answer: We have not considered this issue carefully before, but habitually apply the way of EE definition of AOD to FMF. 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%). After carefully considering your comments, we think that the EE of FMF should not be a function of FMF, so we changed the EE of FMF in this study to +/-0.2, which considered our original absolute error (0.1) in the EE and the uncertainties of AERONET FMF (be of order 0.1 to 0.15, we used a value of 0.1) mentioned in your next comment. The 'EE lines' and 'Within EE' on all the scatter plots were modified.

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.

4. Line 154: Also, when calculating this metric, the uncertainty on AERONET FMF should be accounted for as well (this is dependent on AOD but can also be of order 0.1 to 0.15: this is discussed in some AERONET publications). Unlike AERONET total AOD, AERONET FMF cannot be considered a reference truth because there are non-negligible uncertainties in both the AERONET SDA and SD retrievals.

Answer: As in the previous answer, we changed the EE of FMF to +/-0.2.

5. Lines 220-221: I believe the latest GRASP is version 2.1, not 2.0.6 as stated here. Also, which GRASP product? There are 3 separate GRASP POLDER data sets with different assumptions about aerosol size distribution form. See this paper by Chen et al for more information: https://essd.copernicus.org/preprints/essd-2020-224/. More information should be added to the manuscript, and if possible the analysis should use the latest GRASP version. Including those results in this paper (rather than just citing an older evaluation study) would also help to compare GRASP and the authors' new approach. Right now it is still not clear to me which is better or what the relative benefits are.

Answer: Yes, the latest GRASP is version 2.1 according to the paper by Chen et al., and the GRASP product used in our previous study is from the «high-precision» approach. However, the latest version can be obtained from AERIS/ICARE Data and Services Center (http://www.icare.univ-lille.fr) is version 2.06 (Figure R3), we can only use this older version of the product for processing. We have added the following relevant information in the revised paper:

In our previous research, the accuracy of FMF calculated from the GRASP «high-precision» product was validated.

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).

Chen et al. did not discuss the uncertainty of FMF of GRASP, and we cannot directly compare the two FMF based on their research. But we have added a discussion about the FMF definition as below:

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.

ICARE On-line Data Archive

DATA > PARASOL

File	Description	Size	Last Modified
..			
AC3-HDF.v1.00/			Jan 05 2017 10:10
AC3-HDF.v1.01/			Sep 26 2016 23:01
AC3-HDF/			Sep 26 2016 23:01
AC3-QL.v07.05.v10.11/			Mar 09 2016 09:26
AC3-QL/			Mar 09 2016 09:26
AC3.v07.05.v10.11/			Jul 21 2015 11:22
AC3/	Aerosols over Land - Level 3		Jul 21 2015 11:22
AER-D3.v1.01/			Apr 04 2017 20:48
AER-D3/			Apr 04 2017 20:48
ANCILLAIRE/			Jul 23 2012 18:43
CDR_L2.v01.02/			Jun 22 2015 01:56
CDR_L2/			Jun 22 2015 01:56
CDR_L3.v01.01/			Jun 14 2019 17:36
CDR_L3/			Jun 14 2019 17:36
GRASP-D3-1deg.v2.01/			Mar 31 2018 07:22
GRASP-D3.v1.00/Restricted access			Mar 22 2017 07:17
GRASP-D3.v2.00/			Apr 28 2017 17:03
GRASP-D3.v2.01/			Mar 31 2018 05:47
GRASP-D3.v2.05/Restricted access			Sep 24 2018 15:27
GRASP-D3.v2.06/			Oct 16 2018 03:06
L1_B-HDF.v1.00/			Jan 05 2017 10:10
L1_B-HDF.v1.01/			Sep 16 2016 01:10
L1_B-HDF/			Sep 16 2016 01:10
L1_B-QL.v03.02/			Mar 09 2016 09:28

Figure R3. The list of datasets on ICARE data center

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.

6. Line 243: if possible, more information about the PM2.5 and PM10 surface measurements should be made here. For example is this BAM, filter, or something else?

Answer: Sorry, the National Bureau of Statistics has not described the PM2.5 and PM10 measurement method in the Statistical Yearbook. As far as I know, the environmental monitoring national control station in China uses two methods, the beta ray method and the oscillatory balance method, but it is uncertain which method is used for the specific station.

7. Section 4.1: I do not see much value in showing these two case studies. It's just a couple of maps and text describing them. There isn't really enough context or external data sets brought in to make them interesting. I recommend removing section 4.1 (so section 4.2 would just be called section 4), unless the authors can provide additional material of scientific interest to make the reader care about these examples.

Answer: We accepted your comment and deleted this section.

8. Section 4.2: It would be good to add AOD maps here as well, for additional context

Answer: We accepted your suggestion and finally deleted the original section 4.

9. Figure 1: "Fuction" should say "Function".

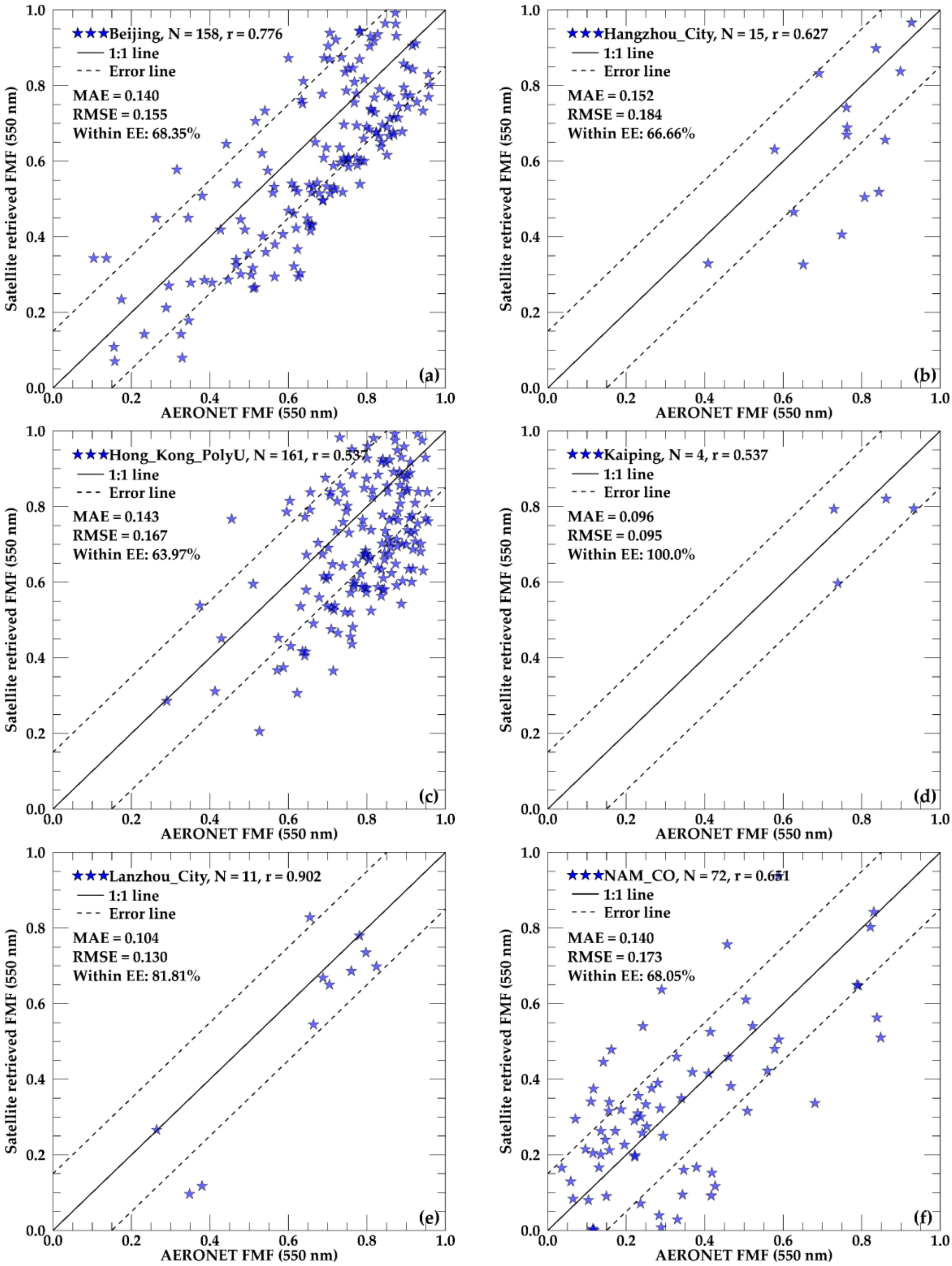
Answer: We have corrected it.

10. Figure 2: I don't know why there is a color bar (FMF) on this figure, given it is only showing site locations on a blank background map

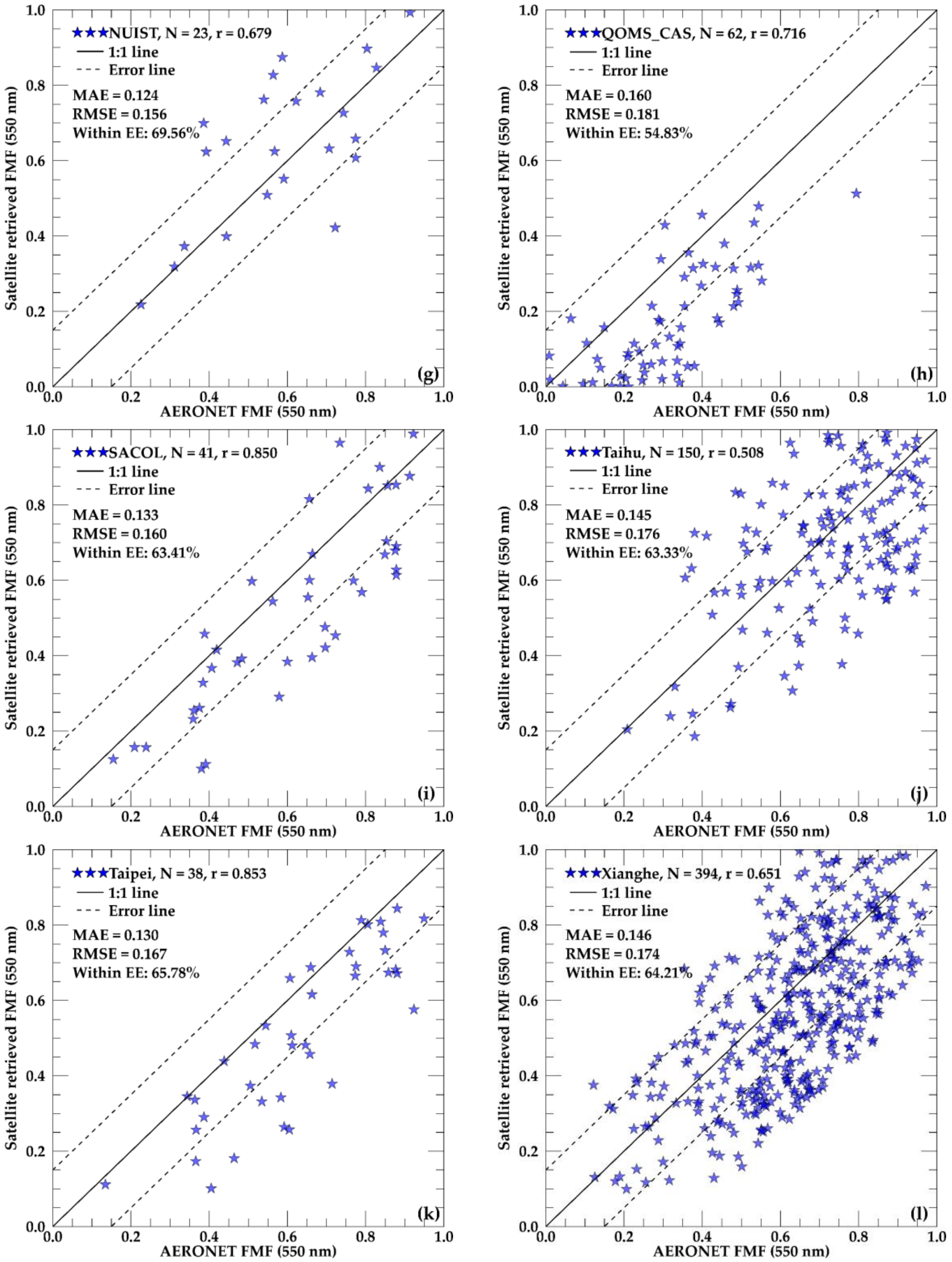
Answer: We have corrected it.

11. Figure 3: These panels should have separate titles or similar (e.g. a, b) to separate them. Also, I would remove the regression lines. I don't think they add anything, and don't think they are appropriate. In some cases the relationships aren't linear (e.g. QOMS_CAS), and the technique is not valid because (1) it is not accounting for uncertainty in the AERONET reference data and (2) the data are constrained by the possible bounds of FMF (i.e. 0-1) meaning that errors cannot be Gaussian and unbiased. Both of these means that the assumptions required for validation are not satisfied.

Answer: The separate titles are in the lower right corner of the figures. We accepted your suggestion and deleted the regression lines. We originally wanted to use a regression line to represent the deviation between the retrieval result and the AERONET result. We did not consider the error of AERONET FMF itself, and did not consider the normal distribution issue, but used it as a true value. The revised figures (Figure R4) are shown as below:



Beijing, N = 158, r = 0.776
1:1 line
Error line
MAE = 0.140
RMSE = 0.155
Within EE: 68.35%
(a)
Hangzhou_City, N = 15, r = 0.627
1:1 line
Error line
MAE = 0.152
RMSE = 0.184
Within EE: 66.66%
(b)
Hong_Kong_PolyU, N = 161, r = 0.537
1:1 line
Error line
MAE = 0.143
RMSE = 0.167
Within EE: 63.97%
(c)
Kaiping, N = 4, r = 0.537
1:1 line
Error line
MAE = 0.096
RMSE = 0.095
Within EE: 100.0%
(d)
Lanzhou_City, N = 11, r = 0.902
1:1 line
Error line
MAE = 0.104
RMSE = 0.130
Within EE: 81.81%
(e)
NAM_CO, N = 72, r = 0.651
1:1 line
Error line
MAE = 0.140
RMSE = 0.173
Within EE: 68.05%
(f)
Satellite retrieved FMF (550 nm)
AERONET FMF (550 nm)
0.0
0.2
0.4
0.6
0.8
1.0


NUIST, N = 23, r = 0.679
1:1 line
Error line
MAE = 0.124
RMSE = 0.156
Within EE: 69.56%
(g)
QOMS_CAS, N = 62, r = 0.716
1:1 line
Error line
MAE = 0.160
RMSE = 0.181
Within EE: 54.83%
(h)
SACOL, N = 41, r = 0.850
1:1 line
Error line
MAE = 0.133
RMSE = 0.160
Within EE: 63.41%
(i)
Taihu, N = 150, r = 0.508
1:1 line
Error line
MAE = 0.145
RMSE = 0.176
Within EE: 63.33%
(j)
Taipei, N = 38, r = 0.853
1:1 line
Error line
MAE = 0.130
RMSE = 0.167
Within EE: 65.78%
(k)
Xianghe, N = 394, r = 0.651
1:1 line
Error line
MAE = 0.146
RMSE = 0.174
Within EE: 64.21%
(l)
Satellite retrieved FMF (550 nm)
AERONET FMF (550 nm)

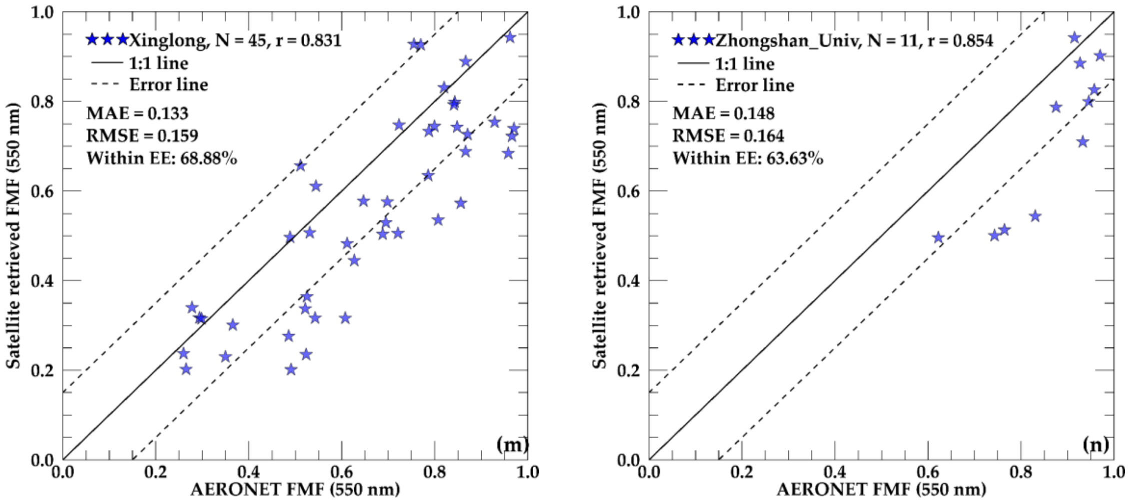


Figure R4. FMF results comparison at 14 AERONET sites. (a) - (n) are the validation results for the Beijing, Hangzhou_city, Hongkong_PolyU, Kaiping, Lanzhou_city, NAM_CO, NUIST, QOMS_CAS, SACOL, Taihu, Taipei, Xianghe, Xinglong, Zhongshan_Univ sites, respectively.

The corresponding 'within EE' in Table 2 has also been modified:

Table R2. FMF validation results of different surface types

Land cover type	N	r	MAE	RMSE	Within EE
Overall result	1186	0.770	0.143	0.170	65.01%
Urban	421	0.733	0.139	0.163	66.98%
Barren	63	0.711	0.158	0.182	55.55%
Grasslands	113	0.777	0.137	0.170	66.37%
Wetlands	150	0.508	0.145	0.176	63.33%
Croplands	394	0.651	0.146	0.174	64.21%
Forests	45	0.831	0.133	0.159	68.88%

12. Figure 4: It would be good to show a second histogram in addition, filtered for points where the AOD is above a certain value (e.g. 0.2?). We would expect that this would be thinner because the sensitivity to FMF should be better when AOD is high. So this would be interesting to see how the width and midpoint of the distribution change.

Answer: In accordance with your opinion, we have added the FMF error distribution result when AOD_f is greater than 0.2 (Figure R5). Comparing the two results, it can be found that after screening, the proportion of FMF error ranging from -0.4 to -0.3 decreased by about 7), and the proportion of FMF error ranging from -0.1 to 0.1 increased by about 6%, which shows that when the AOD is higher, our FMF retrieval method is more sensitive.

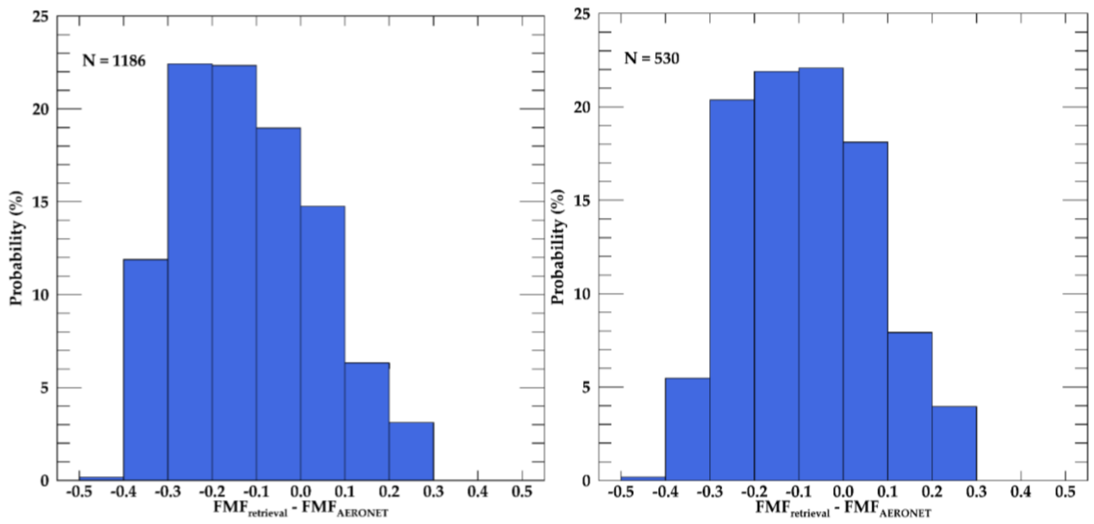


Figure R5. FMF retrieval error distribution results. (a) is for all results, (b) is for the results with AODf greater than 0.2.

13. Figure 5: same comment about regression line as for Figure 3. Also, both data sets have 143 points here: is this for the points where MODIS and POLDER are all matched together? This should be stated in the paper, this was not clear to me.

Answer: We have deleted the regression line, and the revised figure (Figure R6) is shown as below. This is for the points where MODIS and POLDER are all matched together. We have stated it in the revised paper as below:

Figure R6 shows the comparison between the two results and the AERONET ground-based observation results from 2011 to 2013, which is the results where both MODIS and POLDER matching the ground-based observations.

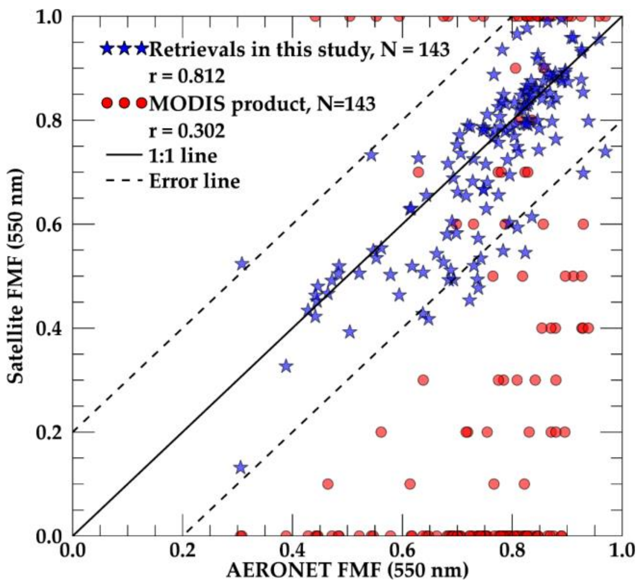


Figure R6. Comparison between the results of this study and MODIS FMF with AERONET

14. Figures 6-8: can we have more than 3 points labeled on the color scale? Also, it would be clearer to combine these together into one 1 figure, possibly with figure 9 as well.

Answer: Now there are 6 points labeled on the color scale, and we combined the original figures 6-9 into one figure (Figure R7) as shown below:

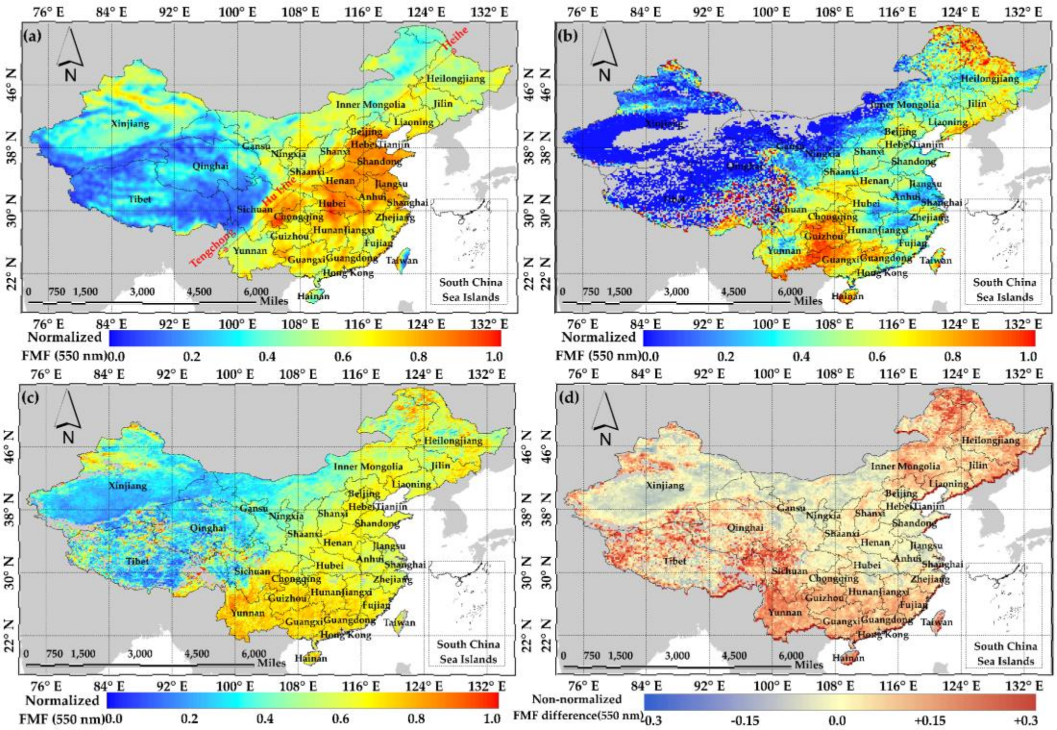


Figure R7. Distribution of FMF of China in 2013 from different data sources. (a) is the normalized results of this study, (b) is the normalized results of MODIS, (c) is the normalized results of GRASP, and (d) is the GRASP results minus the retrieved results (non-normalized).

15. Figure 9: is this FMF difference or normalized FMF difference? This was not clear to me.

Answer: This is non-normalized FMF difference. We have modified the figure and the corresponding description in the paper as shown in the previous answer.

16. Figure 10: same comment about regression line as for Figure 3.

Answer: We have deleted the regression line, and the revised figure (Figure R8) is shown as below:

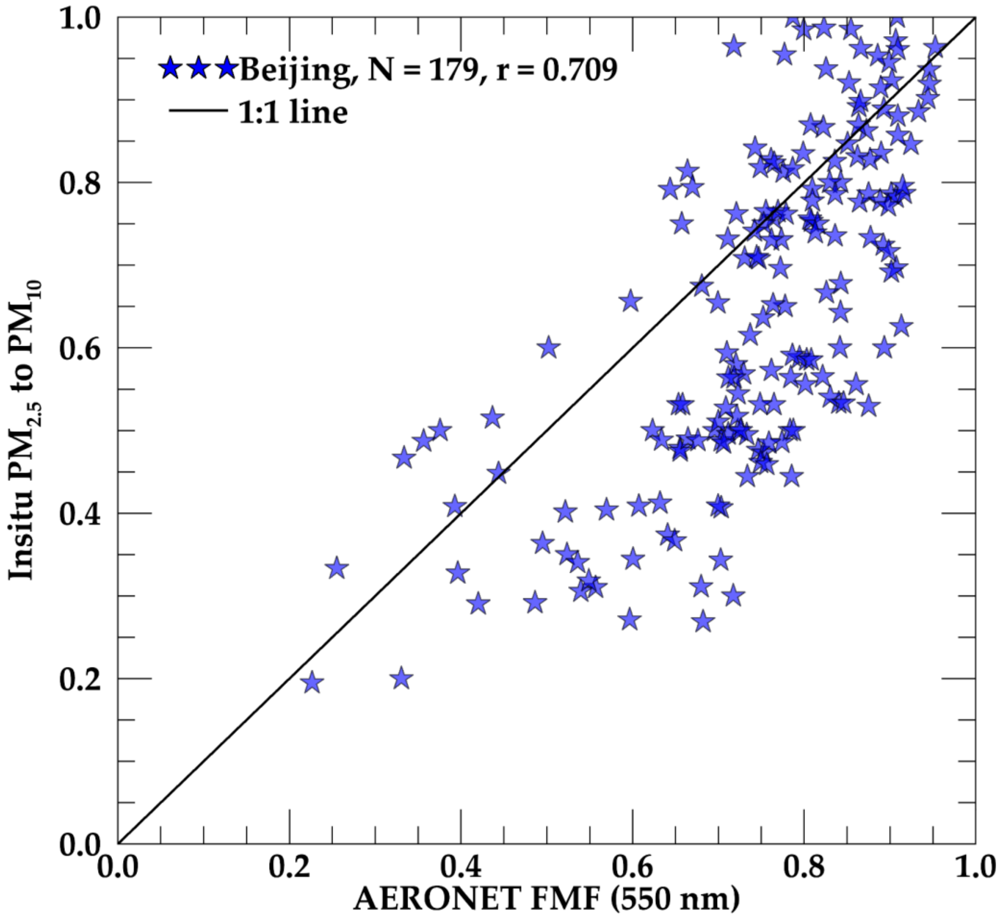


Figure R8. Comparison between the ratio of PM2.5 to PM10 and FMF (hourly average)

17. Figures 15, 16: I don't see much value in these figures and suggest deleting them. The overall spatial distribution looks fairly similar year to year, and I think the seasonal maps in Figure 17 are more useful. Likewise, I am not convinced that the differences in Figure 16 are realistic. It looks like a FMF difference of +0.2 across many parts of China, even remote areas where the aerosol is mostly dust. So in my view it could easily be a calibration drift, as POLDER had no on-board calibration. There is only a paragraph devoted to Figure 16 anyway. If the authors wish to discuss trends, it would make more sense to show also total AOD and fine mode AOD (so we can see which is increasing) and bring in some additional satellite, model, or ground-based data to help verify and understand the mechanisms. If Figures 15 and 16 are removed, then Figure 17 could also be moved earlier in the manuscript, close to where the authors' retrieval method is introduced.

Answer: We accepted your suggestion and considered that the analysis on AOD and FMF may be written as a separate paper, so Figure 17 and related content are moved after the method introduction. In this way, the revised paper does not have the original section 4, the focus of the whole paper is the validation and comparison of the FMF results, and the corresponding paper title is also revised as 'Retrieval of Aerosol Fine-mode Fraction over China from Satellite Multiangle Polarized Observations: Validation and Comparison'.

Responses to reviewer 2

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.

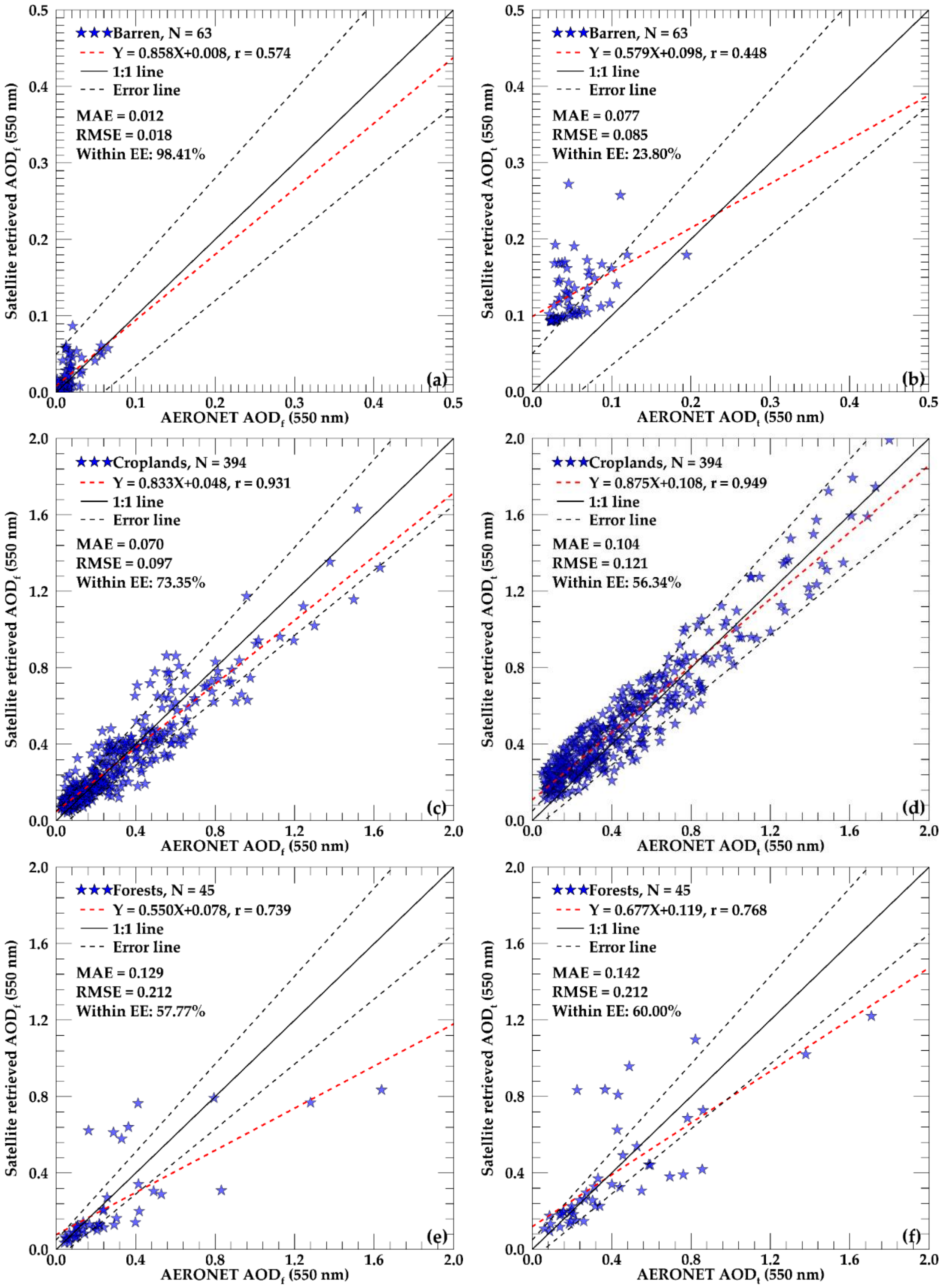
Line 182: the definitions of AERONET FMF and retrieved AODf/AOD are not identical.

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)
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)
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)
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)
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)
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_f (550 nm)
Satellite retrieved AOD_t (550 nm)
AERONET AOD_f (550 nm)
AERONET AOD_t (550 nm)

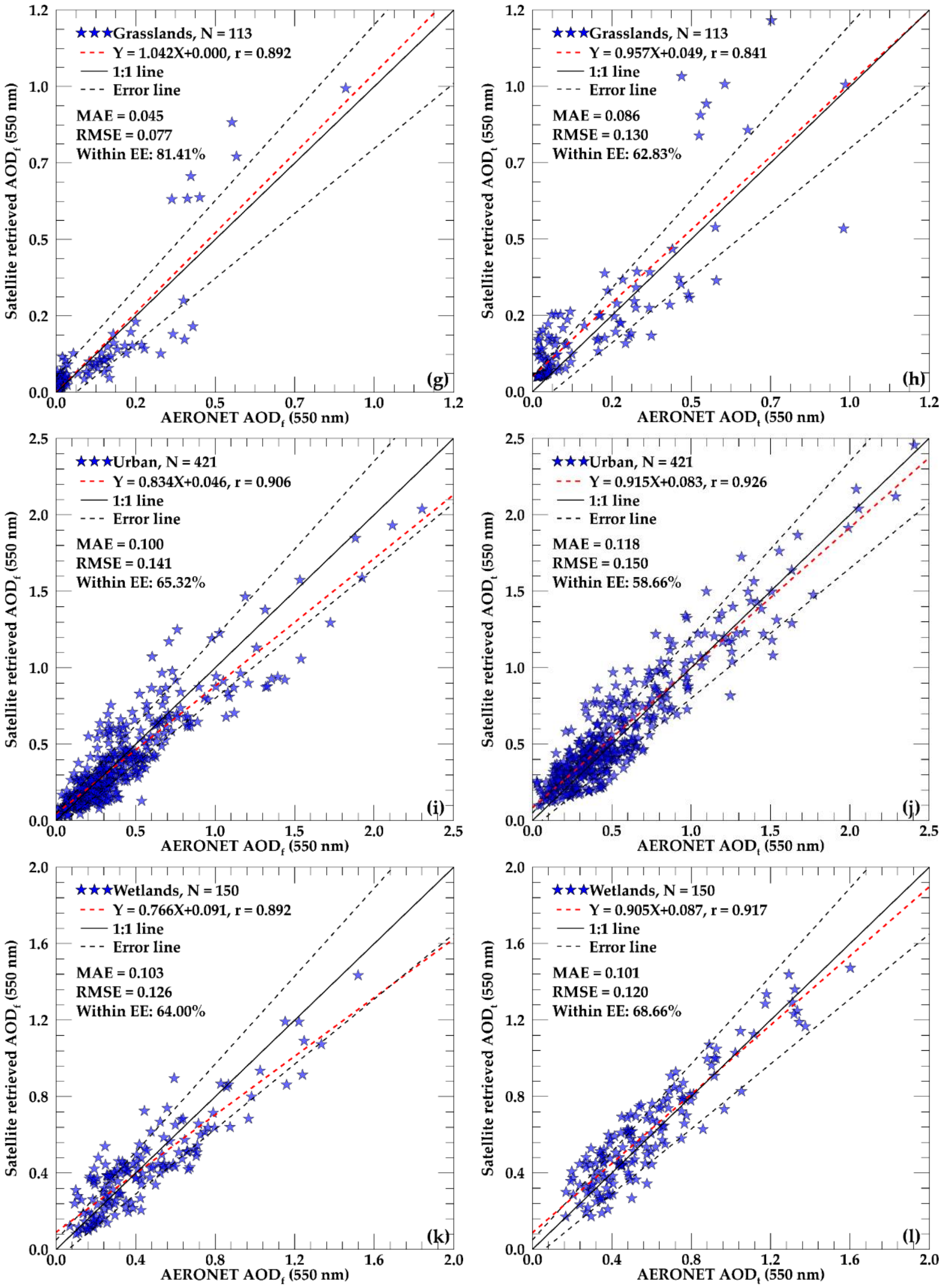


Figure R1. AODs results comparison of 6 surface types. (a), (c), (e), (g), (i), and (k) are the AOD_t validation results for the type of barren, croplands, forests, grasslands, urban, and wetlands, respectively. (b), (d), (f), (h), (j), and (l) are the AOD_f validation results for the type of barren, 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
	AOD_f		0.574	0.006	44.44%	55.56%
Barren	AOD_t	63	0.448	0.111	1.59%	98.41%
	FMF		0.711	-0.144	87.30%	12.70%
	AOD_f		0.931	-0.038	55.84%	44.16%
Croplands	AOD_t	394	0.949	0.077	27.16%	72.84%
	FMF		0.651	-0.064	64.47%	35.53%
	AOD_f		0.739	-0.049	64.44%	35.56%
Forests	AOD_t	45	0.768	-0.019	48.89%	51.11%
	FMF		0.831	-0.102	75.56%	24.44%
	AOD_f		0.892	0.007	38.05%	61.95%
Grasslands	AOD_t	113	0.841	0.061	23.89%	76.11%
	FMF		0.777	-0.033	55.75%	44.25%
	AOD_f		0.906	-0.043	64.61%	35.39%
Urban	AOD_t	421	0.926	0.057	38.72%	61.28%
	FMF		0.733	-0.079	72.45%	27.55%
	AOD_f		0.892	-0.065	69.33%	30.67%
Wetlands	AOD_t	150	0.917	0.048	37.33%	62.67%
	FMF		0.508	-0.031	55.33%	44.67%
	AOD_f		0.868	-0.037	58.68%	41.32%
Overall	AOD_t	1186	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).

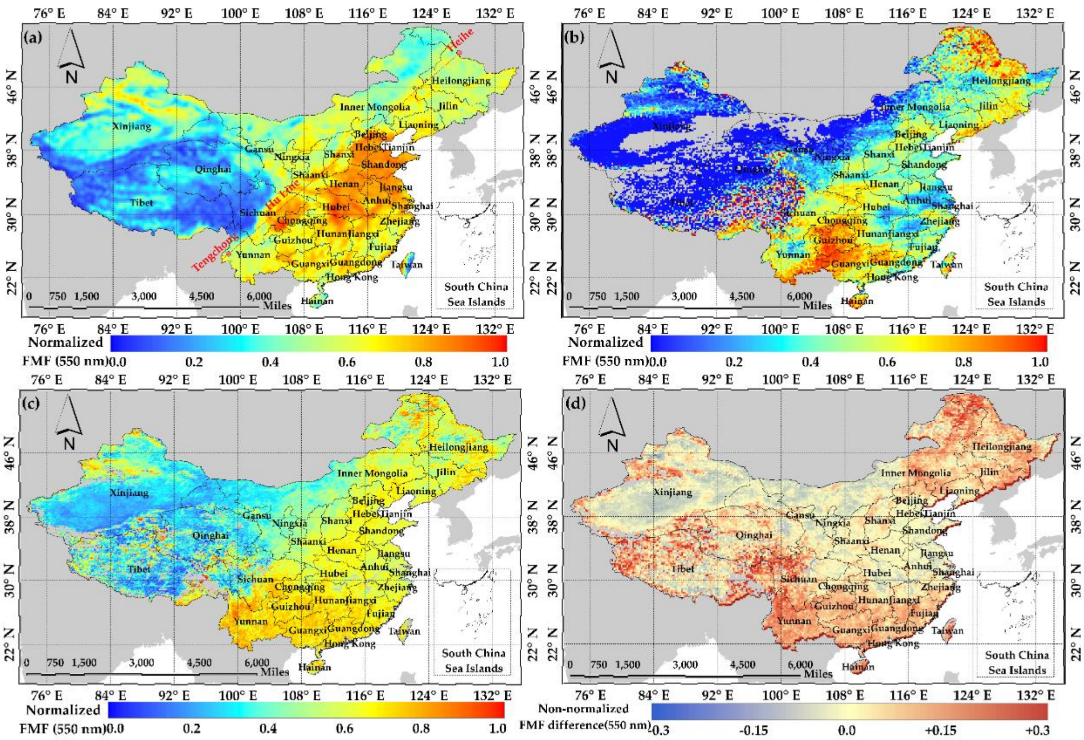


Figure R2. Distribution of FMF of China in 2013 from different sources. (a) is the normalized results of this study (18 km resolution), (b) is the normalized results of MODIS (10 km resolution), (c) is the normalized results of GRASP (6 km resolution), and (d) is the GRASP results minus the retrieved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

Retrieval of Aerosol Fine-mode Fraction over China from Satellite Multiangle Polarized Observations: Validation and ~~Application~~Comparison

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Abstract. The aerosol fine-mode fraction (FMF) is an important optical parameter of aerosols, and the FMF is difficult to accurately retrieve by traditional satellite remote sensing methods. In this study, FMF retrieval was carried out based on the multiangle polarization data of Polarization and Anisotropy of Reflectances for Atmospheric Science coupled with Observations from Lidar (PARASOL), which overcame the shortcomings of the FMF retrieval algorithm in our previous research. In this research, FMF retrieval was carried out in China and compared with the AErosol RObotic NETwork (AERONET) ground-based observation results, Moderate Resolution Imaging Spectroradiometer (MODIS) FMF products, and Generalized Retrieval of Aerosol and Surface Properties (GRASP) FMF results. In addition, ~~application of~~ the FMF retrieval algorithm was ~~carried out~~applied, a new FMF dataset was produced, and the annual and quarterly average results of FMF from 2006 to 2013 were obtained ~~in~~ for all of China. The research results show that the FMF retrieval results of this study are comparable with the AERONET ground-based observation results in China, ~~with~~ and the correlation coefficient (r), mean absolute error (MAE), root mean square error (RMSE), and the proportion of results that fall within the expected error (Within EE) are 0.770, 0.143, 0.170, and ~~60.96~~65.01%, respectively. Compared with the MODIS FMF products, the FMF results of this study are closer to the AERONET ground-based observations. Compared with the FMF results of GRASP, the FMF results of this study are closer to the spatial variation in the ratio of $PM_{2.5}$ to PM_{10} near the ground. ~~The analysis of the annual and seasonal average FMF of China from 2006 to 2013 shows that the FMF high value area in China is mainly maintained in the area east of the "Hu Line", with the highest FMF year being 2013, and the highest FMF season is winter.~~

1 Introduction

Aerosols have a great impact on human production and life and climate change (Kaufman et al., 2002;Huang et al., 2014;Shi et al., 2018). Aerosols have become a research hotspot for scientists from various fields. There are many methods for monitoring aerosols, among which the large-scale coverage of remote sensing technology makes it an effective method for monitoring aerosols. Aerosols produce strong scattering effects in the visible light band (Kokhanovsky et al., 2015). Therefore, in current satellite remote sensing, visible light channels are generally used to observe aerosols, and aerosol information can be obtained on a global scale. At present, in the field of atmospheric environmental research, aerosol optical depth (AOD) products produced by traditional satellite remote sensing platforms, such as Moderate Resolution Imaging Spectroradiometer (MODIS), are the most commonly used (Bellouin et al., 2005;Lee et al., 2011;Xie et al., 2015;Zhao et al., 2017;Zhang et al., 2020). Related scholars have carried out many AOD retrieval studies on traditional scalar observation platforms, which can achieve high-precision retrievals and retrieval of AOD (Li et al., 2013;Kim et al., 2014;Zhang et al., 2014;Zhong et al., 2017;Ge et al., 2019). However, other new aerosol optical parameters, such as the fine-mode fraction (FMF), ~~have poor retrieval accuracy over land, which is~~are quite different in definition from the ground-based observations~~ results~~ (Remer et al., 2005;Levy et al., 2010), which makes them incomparable. ~~.~~The FMF is a parameter that can reflect the content of human-made aerosols (Bellouin et al., 2005;Kaufman et al., 2005), and application requirements have been put forward in many studies. For example, in the particle remote sensing (PMRS) model based on the pure physical approach proposed by Zhang and Li, FMF is one of the core input parameters that determines the final particle concentration retrieval accuracy (Zhang and Li, 2015;Li et al., 2016). However, the existing publicly released satellite FMF products have poor accuracy, which severely limits the retrieval accuracy of the model.

Multiangle polarization observations are a frontier research direction in the field of aerosol remote sensing. These observations have unique advantages in the retrieval of aerosol parameters. Related information analysis work shows that polarization observations can obtain more aerosol information than scalar observations (Chen et al., 2017a;Chen et al., 2017b;Hou et al., 2018). Therefore, the accurate acquisition of more new aerosol parameters based on multiangle polarization observations is of great significance for both atmospheric environmental research and the development of aerosol basic retrieval algorithms. Although official institutions and some scholars have carried out retrieval studies of aerosol parameters based on multiangle polarization observation platforms, such as POLarization and Directionality of the Earth's Reflectances (POLDER), these studies have their own limitations. For example, the French Laboratoire d'Optique Atmospherique (LOA) only ~~released~~ provided the ~~main product of~~ fine-mode aerosol optical depth (AOD_f) datasets in its operational products over land (Deuzé et al., 2001;Tanré et al., 2011), the total aerosol optical depth (AOD_t) was not provided (Chen et al., 2020).~~.~~ Dubovik et al. proposed an optimized retrieval method for polarization observation platforms that can obtain high-precision aerosol optical parameters (Dubovik et al., 2011). Recently, an operational aerosol product of Generalized Retrieval of Aerosol and Surface Properties (GRASP) based on POLDER data was released (Dubovik et al., 2014), and relevant validation studies show that the product has high retrieval accuracy (Tan et al., 2019;Wei et al., 2020). However, ~~as far as~~with regard to this method~~ is concerned~~, its computational convergence speed is slow, computational resources are consumed, and a large amount of mathematical statistics is involved. Compared with the traditional lookup table (LUT) method, this method is more difficult to implement. Although other scholars are conducting related research(Chen et al., 2018;Frouin et al., 2019;Schuster et al., 2019;Li et al., 2020), it is still seldom used in actual engineering applications. In the research of other scholars on the retrieval of new aerosol parameters based on the LUT method, although the results produced by the algorithm have high retrieval accuracy, these studies generally only focus on a specific area, and the spatial scale is not large (Cheng et al., 2012;Xie et al., 2013;Wang et al., 2015;Qie et al., 2015;Wang et al., 2018). There are also fewer studies on the production of long-term aerosol optical parameter datasets. In 2016, we proposed a method for retrieving FMF based on satellite multiangle scalar and polarization observations (Zhang et al., 2016), mainly based on multiangle scalar observations to obtain AOD_t ~~total aerosol optical depth (AOD_t)~~, and multiangle polarization observations to obtain AOD_f. The ratio of the two is FMF. Compared with the existing MOIDS FMF products, the accuracy of the FMF results obtained by this method is significantly improved, which shows the feasibility of the method. However, there are still some problems that need to be solved if this method is to be applied in large spaces. For example, the empirical parameters of surface reflectance estimation during scalar retrieval vary greatly with region, and high-precision AOD_t retrieval results can only be obtained in specific regions. In polarization retrieval, ~~there is a~~the problem of a low AOD_f retrieval value for high aerosol loading exists (Chen et al., 2015;Zhang et al., 2018). In response to these problems, we have also carried out follow-up research work, made certain improvements to the above problems and have achieved more accurate AOD_t and AOD_f in a large space (Zhang et al., 2017;Zhang et al., 2018). Then, in theory, it is possible to achieve the goal of FMF in a large space. Although Yan et al. achieved high-precision FMF retrieval based on the LUT-SDA method (Yan et al., 2017;Yan et al., 2019), their method is mainly oriented to traditional multispectral scalar sensors. To apply this method to multiangle polarization sensors, it is necessary to perform a series of algorithm adjustments. In previous research, we have achieved high-precision retrieval of AOD_t and AOD_f in a large space. The retrieval method and results can be directly used to obtain FMF without additional~~, and no more~~ algorithmic adjustments~~ are needed~~. This paper is mainly based on the POLDER-3 multiangle polarization sensor on the Polarization and Anisotropy of Reflectances for Atmospheric Science coupled with Observations from a Lidar (PARASOL) satellite and the existing research foundation, and it carried out the retrieval and validation of the FMF in the land area of China. The second chapter of the ~~thesis~~ study briefly introduces the FMF retrieval algorithm based on multiangle polarization observation, AErosol RObotic NETwork (AERONET) data and the data validation method.~~;~~ T~~t~~he third chapter mainly compares the retrieval results based on the AERONET ground-based observation data. At the same time, it was also compared with the operational aerosol products of MODIS and GRASP. ~~In Chapter 4, a case study of FMF retrieval is conducted. In this chapter, we also produced a new FMF data set based on the FMF retrieval algorithm of this research. The results of the FMF temporal and spatial distribution over land in China from 2006 to 2013 are obtained.~~ Chapter 4~~5~~ summarizes the full text and proposes future work prospects.

2 Methodology

2.1 Introduction to the FMF retrieval method

The technical framework of FMF retrieval in this research is shown in Figure 1. Overall, the FMF retrieval in this study consists of two parts, namely, using the multiangle scalar and polarization data of POLDER-3 to obtain AOD_t and AOD_f, and the final ratio of the two is FMF. This method is the same as the retrieval method proposed in our 2016 study (Zhang et al., 2016).

However, our previous method is limited by semiempirical parameters on the surface and can only obtain better FMF results at the urban scale. To obtain stable and accurate results in a large space, we have made major changes to the retrieval methods of AOD_t and AOD_f. For the specific retrieval method, please refer to the research we published in 2017 and 2018; here, only a brief introduction is given.

For the retrieval of AOD_t, we introduced the empirical orthogonal function (EOF) to estimate the surface reflection contribution under multiangle observations to solve the regional limitation of the semiempirical parameters of the surface in the original method. Subsequently, this is combined with the retrieval lookup table and substituted into the forward model for simulation calculation, and finally, AOD_t can be obtained through the cost function. The correlation coefficient (r) and root mean square error (RMSE) between the obtained AOD_t and AERONET ground-based observations are 0.891 and 0.097, respectively. The EOF method has previously been used for the retrieval of land aerosols on Multi-angle Imaging Spectro Radiometer (MISR); we transplanted this method to POLDER based on the MISR approach. For more details~~ about the EOF method~~, please refer to our 2017 study (Zhang et al., 2017).

For the retrieval of AOD_f, our research and that of other scholars have shown that the AOD_f results obtained by using the ~~official~~ operational LOA algorithm have a certain deviation compared with ground-based observations. To improve the retrieval accuracy of AOD_f, we proposed the Grouped Residual Error Sorting (GRES) method in 2018 to solve the problem of an inaccurate evaluation function caused by error accumulation under multiangle observation. Based on this method, combined with a bidirectional polarized surface reflectance (BPDF) model to estimate the polarized surface reflectance (Nadal and Bréon, 1999), we have obtained higher-precision AOD_f results in eastern China, and the r and RMSE between the results and the AERONET ground-based observations are 0.931 and 0.042, respectively. More method details can be found in our research published in 2018 (Zhang et al., 2018).

Based on the new retrieval method, we have obtained higher-precision AOD_t and AOD_f retrieval results on a large spatial scale, which also provides the possibility of obtaining accurate FMF results on a large spatial scale. Figure ~~17~~ 2 shows the seasonal average spatial distribution results of FMF in China from 2006 to 2013 obtained by this study. In the figure, spring is from March to May, summer is from June to August, autumn is from September to November, and winter is from December to February. As seen from the figure, for the east area of the "Hu Line", the overall FMF reached its highest value in winter, mainly concentrated in the range of 0.7-0.8; the FMF of southern China still has a relatively high value in the spring, and the overall value is approximately 0.6, while in North China, the plain area is lower, generally between 0.4-0.5; the North China Plain in summer is similar to that in spring;~~,~~ but there is a significant decline in southern China, where the value is generally between 0.3-0.5; and in autumn, the overall value begins to rise, with a~~the~~ value ~~is~~ of approximately 0.6. The Sichuan-Chongqing economic zone maintains a relatively high value in all four seasons, and the value in some areas in winter is close to 0.8, while ~~;~~ the three northeastern provinces also have high values in winter, ~~and~~ with the overall value~~ is~~ between 0.4-0.7. For the area west of the “Hu line”, the northern Xinjiang area is higher in autumn and winter, and it can reach 0.7 in some areas in winter, and the southern Xinjiang area also shows a significant increase in winter, with some high values close to 0.6~~;~~, whereas the Qinghai-Tibet Plateau maintains a low value in all seasons, and the value is mainly concentrated between 0.1-0.3.

Next, we will~~ obtain FMF based on the AOD$_t$ and AOD$_f$ retrieved by the new method,~~ validate the FMF retrieval results based on the AERONET ground-based observation results~~ and further obtain the FMF temporal and spatial distribution results over terrestrial China~~. Note that since the EOFs during the AOD$_t$ retrieval need to be constructed with the observation results of the POLDER 3*3 window, the resolution of the final FMF retrieval result is also the size of the POLDER 3*3 window (approximately 18 km).

2.2 AERONET data

At present, aerosol ground-based products of AERONET have been developed to version V3, and the data of version V2 are no longer available for download. Among these products, there are two products that can be used to validate the results of satellite FMF retrieval: one is the FMF product based on the spectral deconvolution (SDA) method (O'Neill et al., 2001a;O'Neill et al., 2001b;O'Neill et al., 2003), and the other is based on the size distribution (SD) retrieval product (Dubovik and King, 2000). Generally, SDA products can provide more FMF ground-based results. At present, most ground-based stations in China provide SDA products with level 2.0 data quality. Therefore, SDA products are the first choice for FMF comparison in this study. However, it is worth pointing out that the Beijing site lacks the SDA product with level 2.0 data quality, so we used the SD product instead. Finally, this study selected the level 2.0 products of 16 AERONET sites in China during 2006-2013 (POLDER on-orbit time) to validate the FMF retrieval results of this study. The specific spatial locations of AERONET sites are shown in Figure ~~2~~3, and the specific site information is shown in Table 1. However, note that not all AERONET sites have long-term observational data. The sites with long-term observational data are the Beijing, Xianghe, Taihu, and Hong_Kong_PolyU sites.

The FMF retrieved in this study is the FMF at 550 nm. Neither the SDA product nor the SD product directly provides the FMF result at this wavelength. Therefore, the AERONET FMF needs to be wavelength converted. For SDA products, the products include AOD$_t$~~,~~ and AOD$_f$ at 500 nm and the corresponding Ångström~~Angstrom~~ Exponent (AE), so the FMF of SDA products can be converted to 550 nm by Eq. (1):

$$FMF_{550,\ SDA} = \frac{\tau_f^{500}\cdot(500/550)^{\alpha_f}}{\tau_t^{500}\cdot(500/550)^{\alpha_t}}, \quad (1)$$

where $FMF_{550,\ SDA}$ is the FMF of the SDA product at 550 nm after conversion, τ_f^{500} is the AOD$_f$ at 500 nm, τ_t^{500} is the AOD$_t$ at 500 nm, α_f is the fine-mode AE, and α_t is the coarse and fine-mode AE.

The SD products provide AOD$_t$ and AOD$_f$ at 440 nm and 675 nm, respectively. Eq. (2)- Eq. (4) can be used to obtain FMF results at 550 nm:

$$\alpha_t = -\frac{ln\,(\tau_t^{675}/\tau_t^{440})}{ln\,(675/440)}, \quad (2)$$

$$\alpha_f = -\frac{ln\,(\tau_f^{675}/\tau_f^{440})}{ln\,(675/440)}, \quad (3)$$

$$FMF_{550,SD} = \frac{\tau_f^{440}\cdot(440/550)^{\alpha_f}}{\tau_t^{440}\cdot(440/550)^{\alpha_t}}, \quad (4)$$

where $FMF_{550,SD}$ is the SD product FMF at 550 nm after conversion, τ_f^{675} is AOD_f at 675 nm, τ_t^{675} is AODt at 675 nm, τ_f^{440} is AOD_f at 675 nm, and τ_t^{440} is AODt at 675 nm.

2.3 Validation method

In this study, the average value of ground-based observation results within ±30 min of the satellite's transit was used for comparison with the satellite retrieval results. 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). Note that when the retrieved AOD_f is greater than the retrieved AODt, 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%.

The statistical indicators used in the ~~verification~~ validation include the correlation coefficient (r), mean absolute error (MAE), bias, RMSE, and expected error (EE). The specific statistical evaluation index definitions are shown in Eq. (5)-Eq. (~~8~~10):

$$r = \frac{Cov(FMF_{retrieval}, FMF_{AERONET})}{\sqrt{D(FMF_{retrieval})}\sqrt{D(FMF_{AERONET})}}, \quad (5)$$

$$MAE = \frac{1}{n}\sum_{i=1}^{n}\left|FMF_{i,retrieval} - FMF_{i,AERONET}\right|, \quad (6)$$

$$Bias = \frac{1}{n}\sum_{i=1}^{n}(FMF_{i,retrieval} - FMF_{i,AERONET}), \quad (7)$$

$$RMSE = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(FMF_{i,retrieval} - FMF_{i,AERONET})^2},$$

(~~7~~8)

EE_{FMF} ~~EE~~ $= \pm 0.$~~1~~2 ~~$\pm 0.1 \times FMF_{AERONET}$~~,

(~~8~~9)

$$EE_{AOD} = \pm 0.05 \pm 0.15 AOD_{AERONET}, \quad (~~9~~10)$$

where *Cov()* represents the covariance, *D()* represents the variance, $FMF_{retrieval}$ represents the FMF retrieval value, $FMF_{AERONET}$ represents the value of AERONET FMF, $AOD_{AERONET}$ represents the value of AERONET AOD, i is the matched data points, and n is the number of validation points.

3 Validation and comparison

3.1 ~~Introduction to the FMF retrieval method~~Validation against AERONET ground-based data

Figure ~~3~~ 4 is a scatter plot of the comparison between the retrieved and AERONET ground-based FMFs. Figures ~~3~~4(a) to ~~3~~4(n) list the verification results at the corresponding sites where the number of matching results is greater than 2. The figure shows that the FMF results obtained in this study have an overall high correlation with the AERONET ground-based observations. Among the 14 AERONET sites, r is between 0.508 (Taihu site) and 0.902 (Lanzhou City site). The ranges of MAE and RMSE are 0.096 (Hangzhou_City site) to 0.160 (QOMS_CAS site) and 0.095 (Hangzhou_City site) to 0.184 (QOMS_CAS site). Except for the QOMS_CAS site, the proportion of results that fell within the EE accounted for approximately ~~60~~65%. The statistical indicators of the QOMS_CAS site are all poor. The specific reason is that the site is located at the southern edge of the Qinghai-Tibet Plateau. It is a high-altitude site and has very little aerosol content. In the AERONET SDA products of 2009-2013, the 5-year average values of AOD_t and AOD_f (500 nm) are only 0.052 and 0.038, respectively. Under the combined influence of the aerosol model and the surface reflectance estimation error in the retrieval process, it is difficult to accurately retrieve a low AOD value for satellite observations, resulting in a large deviation of FMF at this site.

We have counted the FMF validation results of different surface types, and the specific information is shown in Table 2. The r, MAE, and RMSE at all sites in this study are 0.770, 0.143, and 0.170, respectively, and Within EE is 6~~0.96~~5.01%, again indicating that the FMF satellite retrieval results of this study are comparable with the ground-based observation results. All the validation results of this study cover six surface types: urban, barren, grasslands, wetlands, croplands, and forests. Overall, since the validation data of the barren type mainly come from the QOMS_CAS site, the validation results at this surface type are poor. Although the r at the other five surface types has a certain change, it is 0.508 (barren)-0.831 (forests)), but in terms of the three indicators of MAE, RMSE and Within EE, the differences in the five surface types are relatively small, especially Within EE, which is concentrated at approximately ~~60~~65%, similar to the site-by-site results. The ~~errors~~ uncertainty of the FMF retrieval results in this study are relatively stable at these five surface types.

We further counted the error distribution of the FMF retrieval results, and the statistical results are shown in Figure ~~4~~5(a). The figure shows that the FMF error of this research is mainly distributed between -0.3 and 0.3. This part of the data accounts for approximately 86%, but the part less than the AERONET ground-based FMF observation value accounts for approximately 75%, indicating that the retrieval result of this study is lower than that of the ground-based observations. ~~The specific reason needs to be analysed from the FMF retrieval method of this study. The FMF in this study is obtained from the ratio of AOD_f to AOD_t, and the retrieval accuracy of the two parameters directly determines the retrieval accuracy of FMF. Therefore, we compared the retrieved AODs with those of the ground-based data in 2013, and the statistical results are shown in Table 3. The table shows that the mean errors between the AOD_f and AOD_t of our retrieval and the ground-based results are -0.039 and 0.043, respectively, indicating that the AOD_f retrieval result has a negative offset, and the AOD_t retrieval result has a positive offset, that is, the numerator is small and the denominator is large, eventually leading to a small FMF.~~We further screened out the points with AOD_f greater than 0.2, and the corresponding FMF error distribution results are shown in Figure 5(b).

Comparing the two figures, it can be found that after screening, the proportion of FMF error ranging from -0.4 to -0.3 decreased by ~~about~~approximately 7%, and the proportion of FMF error ranging from -0.1 to 0.1 increased by ~~about~~approximately 6%, which shows that when the AOD is higher, our FMF retrieval method is more sensitive.

Since our FMF is obtained from the ratio of AOD_f and AOD_t retrieval results~~The specific reason needs to be analysed from the FMF retrieval method of this study. The FMF in this study is obtained from the ratio of AOD_f to AOD_t~~, and the retrieval accuracy of the two parameters directly determines the retrieval accuracy of FMF~~. Therefore~~, we further compared the retrieved AODs at the six different surface types with those of the ground-based data ~~in~~from 2006 to 2013, and the statistical results are shown in Figure 6 and Table 3. It can be seen from Figure 6 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 analys~~z~~e 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. ~~The t~~Table 3 shows that the ~~mean errors~~bias ~~between~~ of the retrieved ~~the~~ AOD_f and AOD_t at the six different surface types. It can be seen from Table 3 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~~of our retrieval and the ground-based results are -0.039 and 0.043, respectively.~~. 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 ~~offset~~bias, and the AOD_t retrieval result has a positive ~~offset~~bias, that is, the numerator is small and the denominator is large, eventually leading to a ~~small~~negative bias of FMF.

3.2 Comparison with MODIS products

MODIS aerosol products also include FMF data sets, but this FMF has a different definition. In fact, the FMF of MODIS refers to the 'fine model fraction', which is the proportion of bimodal fine-dominated aerosol model, but not pure fine mode (Levy et al., 2007). Because the FMF results obtained by MODIS are different in definition from the ground-based results (Levy et al., 2009), the retrieval results are quite different from the ground-based observation results, which limits the research that depends on the FMF parameter. We compared the retrieved and MODIS FMF with the AERONET ground-based observations to further evaluate the significance of our results. The MODIS FMF results were derived from the MYD04 product of collection 6.1. Figure ~~5~~ 7 shows the comparison between the two results and the AERONET ground-based observation results from 2011 to 2013, which is the results where both MODIS and POLDER matching the ground-based observations. As seen from the figure, compared with ground-based observations, the r of FMF obtained in this study is 0.812, while that of MODIS is 0.302.

The correlation coefficient of the results obtained in this study is much higher than that of MODIS. At the same time, notice that there are many 0 values in the MODIS results. These 0 values are not meaningless but correspond to the situation where there ~~are~~ is no the fine~~ particle~~-dominated aerosol model~~s in the MODIS product definition~~. ~~Judging from the comparison results, these 0 values have large deviations from the ground-based observation results, and the results of this study are closer to the ground-based observations.~~

More statistical results of the two are shown in Table 4. The table shows that the FMF results obtained in this study have an MAE of 0.072, an RMSE of 0.102, and a Within EE of ~~79.72~~87.41%,~~;~~ ~~the~~ whereas results of MODIS have an MAE of 0.512, RMSE of 0.574, and Within EE of 1~~2.59~~9.58%. The statistical indicators of the FMF results obtained by our study are ~~greatly improved~~closer to the ground-based observations than the~~ compared with the~~ MODIS results. ~~But~~Nevertheless, note that this does not mean that the FMF of MODIS has a large deviation. As mentioned above, there is a difference in definition between the FMF of MODIS and the ground-based observations~~,~~; consequently, it~~it~~ is difficult to obtain the true deviation of MODIS FMF based on ground-based observations.

Figure ~~6 and Figure 7~~8(a) to 8(b) show the spatial distribution map of the average annual FMF (550 nm) of China in 2013 obtained by this study and the MODIS product. To facilitate the comparison of the differences in the spatial distribution trends of the two, the two results are normalized, meaning they are divided by the maximum value in the respective FMF image. The figures show~~s~~ that the results obtained in this study can better reflect the differences in the level of urbanization in China and are more in line with the "Hu Line", reflecting China's population density. That is, in the area to the east of the "Hu Line", the value of the FMF is higher, and the North China Plain, Sichuan-Chongqing Economic Zone, Pearl River Delta, and Yangtze River Delta are extremely high value areas, while in the area to the west of the "Hu Line", the FMF value is small, the high-value area is mainly in the northern Xinjiang region, while the value in the Qinghai-Tibet Plateau is generally low. The results of MODIS are quite different from the results of this study. The MODIS results show that the regions with the highest FMF are Guizhou, Guangxi, Yunnan, and Hainan. The Three Northeast Provinces and the central mountainous areas of Taiwan also have high values. For the North China Plain, Sichuan-Chongqing Economic Zone, and Pearl River Delta, the results are somewhat similar to this study, while the Yangtze River Delta is a low-value area.

3.3 Comparison with GRASP products

In our previous research, the accuracy of FMF calculated from the GRASP «high-precision» product was validated (Wei et al., 2020). The results of comparison with 8 SONET (Sun-sky radiometer Observation NETwork) sites show that the r between GRASP FMF and ground-based observations is 0.77, and Within EE is 62.35%, which is similar to the results of this study in Section 3.1. However, by comparing the spatial distribution results of the two, we found some differences. ~~We processed the latest V2.06 version of GRASP aerosol products.~~ 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). Figure 8(c) shows the annual averaged FMF spatial distribution of GRASP in 2013 (also normalized). Compared with Figure ~~6~~8(a), we can see certain differences. The relatively high~~-~~value area of GRASP results is mainly in southern China. We subtracted the results of this study from the average GRASP FMF results and obtained the non-normalized numerical difference between the two, as shown in Figure ~~9~~8(d). The figure shows that the difference between the two in the North China Plain and the southern Xinjiang region is relatively small. The largest differences are mainly concentrated in the southern and northeastern China and Qinghai-Tibet Plateau regions. The GRASP results in these areas are greater than our results, and a small number of pixels can be larger than 0.3. However, these areas lacked publicly available sunphotometer observations in 2013 and ~~before~~prior years. The PARASOL ended its exploration mission in October 2013, and it is impossible to compare the subsequent time periods, so it is difficult to directly compare with ground-based observations to illustrate the correctness of the spatial distribution of the two.

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. ~~But~~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.

~~In order to~~To show that the spatial distribution of the FMF in this study is reasonable,~~In this study,~~ the ground PM2.5 and PM10 in situ results were compared with the ground-based FMF results. It is expected that the ratio of $PM_{2.5}$ to PM_{10} can be used to analyse the correctness of this study, as well as the GRASP FMF results in the spatial distribution trend. We selected the 2015 Beijing Olympic Sports Center monitoring site (116.407°E, 40.003°N, straight-line distance of less than 4 km), which was the closest to the AERONET Beijing site, and compared the hourly averaged results of the ratio of $PM_{2.5}$ to PM_{10} with the FMF results. Although the definitions of the two are quite different, the ratio of $PM_{2.5}$ to PM_{10} is actually a parameter of particulate matter near the ground, while FMF is actually a parameter of the atmospheric column of aerosols, but the comparison results of the two (Figure ~~10~~9) show that there is a correlation between the ratio of $PM_{2.5}$ to PM_{10} and FMF, and the r is 0.709. This result may be because aerosols are mainly distributed near the ground, and $PM_{2.5}$ and PM_{10} can represent different particle modes. Ultimately~~In the end~~, the actual difference between the two parameters is smaller. Since the ratio of $PM_{2.5}$ to PM_{10} is comparable to the ground-based FMF results, if there ~~are~~ were more in situ data, it ~~can~~ could indirectly verify the spatial distribution trend of this study and the GRASP results.

Due to the lack of in situ data for particulate matter in China in 2013, this study can only be based on the 2013 environmental protection key city air in the China Statistical Yearbook (http://www.stats.gov.cn/tjsj/ndsj/). The annual average value of air quality is used for limited analysis. We extracted the FMF retrieval results and GRASP results of the corresponding 47 cities in the statistical yearbook and calculated the annual average FMF of each city for comparison with the ratio of the annual average $PM_{2.5}$ to PM_{10} of each city. The spatial distribution of the administrative regions of these 47 cities is shown in Figure ~~11~~10. These cities cover most of China's provinces and have a wider spatial distribution range than the AERONET sites in Figure ~~2~~1. The comparison results in Figure ~~12~~ 11 show that although the annual average FMF results of this study in each city are lower than the annual average results of the ratio of $PM_{2.5}$ to PM_{10}, the change trend of the FMF results of this study is better than the results of GRASP FMF. The r between the FMF of this study and the ratio of $PM_{2.5}$ to PM_{10} is 0.778, while GRASP is 0.472, which can provide evidence for the correctness of the FMF results of this study in the spatial distribution. The low FMF results in this study are related to the calculation methods of the annual average values of $PM_{2.5}$ and PM_{10} in each city. Generally, most of the in situ monitoring sites for particulate matter in each city are distributed in urban areas, and the number of sites distributed in rural areas is small (for example, 9 of the 12 state-controlled sites in Beijing are in urban areas). When calculating the average FMF of a city, one pixel may contain the results of multiple monitoring stations in place, which makes it difficult to achieve accurate spatial location matching. To facilitate data processing, all pixels within the urban administrative boundary are directly used to calculate the average value, and the large number of FMFs in rural areas is generally lower than that in cities, which ultimately leads to a lower FMF average result.

Based on the validation and comparison results in Sections 3.1 to 3.3, this research has obtained FMF satellite retrieval results with good accuracy in China, which proves the reliability and stability of the retrieval method. Compared with the MODIS FMF products, the r, MAE, RMSE and Within EE of the results of this study are all higher than the results of MODIS. Compared with the GRASP FMF, the results of this study are closer to the results of the ratio of $PM_{2.5}$ to PM_{10} in terms of the spatial distribution of the entire region of China. The above results all illustrate the effectiveness and advantages of the FMF retrieval method used in this study. Compared with our original FMF retrieval method, which can only be used at the urban area scale, this research has achieved FMF retrieval in a large space. ~~Therefore, we will carry out the practical application of FMF satellite remote sensing retrieval based on the new method.~~

~~4 FMF retrieval application~~

~~4.1 Case study~~

~~4.1.1 A haze case in North China~~

~~Figure 13 contains the retrieval results of a haze pollution incident that occurred in North China on October 5, 2013. The true colour map shows that North China, especially the Beijing-Tianjin-Hebei region, has several smoke-like pixels, which is a typical feature of haze pollution in satellite remote sensing images.~~

~~Regarding the spatial distribution of AOD_t, the AOD_t value (550 nm) in most areas of Beijing-Tianjin-Hebei exceeded 1.0, and the actual maximum value could reach approximately 2.0. It was the most severely polluted area of the day. There was also a haze distribution in other surrounding areas, such as eastern Shanxi and northern Shandong, and most areas of Henan have areas with an AOD_t value of approximately 0.75-1.0. In addition, there are some areas with an AOD_t value of approximately 0.75 in Anhui and Jiangsu, which fully demonstrates that haze pollution in China is characterized by a large continuous distribution. In southern China, only the Pearl River Delta and southern Taiwan have areas with a value of~~

approximately 0.5. The other regions, such as Hubei, Hunan, Jiangxi, Guangdong and Fujian, have low AOD_t values, and most of the values are concentrated at approximately 0.2, which reflects the cleaner air conditions in South China.

Regarding the spatial distribution of AOD_f, the values (550 nm) in most areas of Beijing-Tianjin-Hebei also exceed 1.0, the actual maximum value can reach approximately 1.6, and the overall spatial distribution is similar to the AOD_t distribution. For South China, the value of AOD_f is also relatively small, most of which is concentrated at approximately 0.15, and the values in the Pearl River Delta and Taiwan are larger, with a value of approximately 0.4.

The spatial distribution of FMF is quite different from the distributions of AOD_t and AOD_f, reflecting that FMF is another observation dimension in aerosol optical properties. The FMF value (550 nm) in most regions of China is concentrated in the range of 0.6-0.8, and the FMF in most parts of South China also reached this level. This result shows that although the air qualities are quite different, the FMF values are close. It also shows that most of the eastern part of China was dominated by fine-mode aerosols, and only parts of Hubei, Hunan, Jiangxi, and Fujian were dominated by coarse-mode aerosols (with FMF values of approximately 0.25).

4.1.2 Case of Sand and Dust in North China

Figure 14 is the retrieval result of a dust pollution incident in North China on March 9, 2013. The true colour image shows that, except for the Beijing area, which is covered by clouds, the other regions of North China are covered by a large number of brown pixels, which is a typical feature of dust pollution in satellite remote sensing images.

Regarding the spatial distribution of AOD_t, the AOD_t value (550 nm) in most areas of the North China Plain is concentrated in the range of 1.2-1.6, and the actual maximum value reaches approximately 2.0, indicating that there is serious dust pollution in most areas of North China. The overall value of Inner Mongolia is relatively small, concentrated at approximately 0.4. Hubei, Anhui, Jiangxi and other regions also have high value areas with AOD_t values of 1.2-1.6, but from the true colour image, these areas are covered by a large number of smoke-like pixels. The high values in these areas are caused by haze pollution; the AOD_t in South China is generally low, mostly below 0.25.

The spatial distribution of AOD_f shows a different trend from AOD_t. The AOD_f (550 nm) of most parts of North China is less than 0.2; Henan, Shandong and other places have areas greater than 0.5; the AOD_f in central China is concentrated between 0.5-1.2, and Hubei has the highest value of 1.2; the overall spatial distribution of South China is similar to that of AOD_t, and the value is also low, generally below 0.3.

Regarding the spatial distribution of FMF, the overall trend is again different from the spatial distribution of the AOD_t and AOD_f. The FMF value (550 nm) in most areas of North China is concentrated in the range of 0.1-0.2, showing that typical coarse-mode aerosols are dominant; notice that there is a transitional area of FMF at the junction of Hebei, Shandong, and Henan, the value varies between 0.1-0.5, which reflects to a certain extent that these areas are affected by dust and haze, and the composition of aerosols is complex. Central China has a higher FMF value, generally above 0.7, reflecting the dominance of fine-mode aerosols. Note that in Jiangsu, Guangdong, Fujian and other places, although the values of AOD_t and AOD_f are both low, the FMF values are still high, greater overall than 0.75, showing a strong trend of fine-mode aerosols dominating.

4.2 Spatiotemporal distribution results of FMF in China's land area from 2006 to 2013

The retrieval of FMF in all of China's land was performed based on the PARASOL level 1 data from 2006 to 2013. We produced a new FMF dataset of China and obtained the corresponding FMF annual and quarterly average results.

Figure 15 shows the results of the FMF annual average spatial distribution of China from 2006 to 2013. In the results, the FMF spatial change characteristics of China in the 8 years are not obvious; that is, the high value area is always dominated by the area east of the "Hu Line", and the high value area of northern Xinjiang is the most conspicuous in western China. Overall, the FMF in China reached its highest in 2013, but there are certain differences in some regions over time. For example, the value of the North China Plain and Yangtze River Delta region from 2011 to 2013 was higher than that of previous years, and the value was mainly between 0.6-0.7; the Pearl River Delta region had a higher value from 2007 to 2008, with a value of approximately 0.65. The overall level of the other years is approximately 0.55 and is lower than that of the North China Plain; the Sichuan-Chongqing Economic Zone has a relatively high value in 2013, the FMF value of the entire region is between 0.5-0.7 and is mainly between 0.4-0.6 in the other year; the northern Xinjiang region is similar to the North China Plain region and has a higher value from 2011 to 2013, but the overall level is lower than that of the North China Plain, and the high value regions are mainly distributed in the economic belt of the northern slope of the Tianshan Mountains, and the value is mainly approximately 0.5.

Figure 16 shows the change in the annual average FMF of China in 2013 compared with 2006. Overall, the annual average value of FMF in China is on the rise, and the provinces with obvious changes in value are mainly Sichuan, Shaanxi, Henan, Hubei, and Yunnan, with an increasing value of up to 0.2. Tibet, Inner Mongolia, Hunan, Jiangxi, and Guangxi have seen negative changes in the annual average FMF, but the decline is only approximately 0.05, and the FMF in these provinces is still mainly positive.

Figure 17 shows the seasonal average spatial distribution results of FMF in China from 2006 to 2013. In the figure, spring is from March to May, summer is from June to August, autumn is from September to November, and winter is from December to February. As seen from the figure, for the east area of the "Hu Line", the overall FMF reached its highest value in winter, mainly concentrated in the range of 0.7-0.8; the FMF of southern China still has a relatively high value in the spring, and the overall value is approximately 0.6, while in North China, the plain area is lower, generally between 0.4-0.5; the North China Plain in summer is similar to that in spring, but there is a significant decline in southern China, the value is generally between 0.3-0.5; in autumn, the overall value begins to rise, the value is approximately 0.6. The Sichuan-Chongqing economic zone maintains a relatively high value in all four seasons, and the value in some areas in winter is close to 0.8; the three northeastern provinces also have high values in winter, and the overall value is between 0.4-0.7. For the area west of the "Hu line", the northern Xinjiang area is higher in autumn and winter, and it can reach 0.7 in some areas in winter, and the southern Xinjiang area also shows a significant increase in winter, with some high values close to 0.6; the Qinghai-Tibet Plateau maintains a low value in all seasons, and the value is mainly concentrated between 0.1-0.3.

~~5~~4 Summary

In this study, the multiangle polarization data of PARASOL were used to perform FMF retrieval, and the retrieval results were compared with the AERONET ground-based observations, MODIS results, and GRASP results.~~ Based on the FMF retrieval method, the retrieval of air pollution cases in China was carried out, and the results of the FMF temporal and spatial distribution in China from 2006 to 2013 were also obtained.~~ Based on the above work content, the conclusions of this research are described as follows:

(1) There is good agreement between the FMF results obtained in this study and the AERONET ground-based observation results. The overall r, MAE, RMSE, and Within EE between the two are 0.770, 0.143, 0.170, and 6~~0.96~~5.01%, respectively.

(2) The FMF results obtained in this study were more practical than the MODIS FMF products. The r, MAE, RMSE, and Within EE between the FMF results and the ground-based observations are 0.812 versus 0.302, 0.072 versus 0.512, 0.102 versus 0.574, ~~79.72~~87.41% versus 1~~2.59~~9.58%, respectively.

(3) Compared with the GRASP FMF, the FMF results obtained in this study are closer to the ratio of $PM_{2.5}$ to PM_{10} in terms of the spatial distribution trend. Compared with the annual average ratio of $PM_{2.5}$ to PM_{10} in 47 Chinese cities in 2013, the r of this study is 0.778, and GRASP is 0.472.

~~(4) According to the annual and quarterly average FMF results in China from 2006 to 2013, the spatial distribution trend of China's FMF does not change significantly with the year, and the high-value area is mainly maintained in the area east of the "Hu Line". The FMF showed an increasing trend in 2013 compared with that of 2006. The FMF in China has the highest value in winter and the lowest value in summer. The Sichuan-Chongqing economic zone has a relatively high FMF value in all four seasons.~~

The FMF retrieval method in this study has significance for the development of aerosol polarization satellite remote sensing algorithms, and the FMF results obtained in China also have good practical value for application research in the field of atmospheric environments. China has launched the Gaofen-5 (GF-5) satellite equipped with a new multiangle polarization sensor. With the release of GF-5 satellite data in the future, the results of this study can also provide algorithmic support for the application of its multiangle polarization sensor in the field of atmospheric environmental monitoring and are expected to produce subsequent FMF datasets. However, there are some shortcomings in this research. For example, the retrieval of FMF still depends on the accuracy of the two parameters AOD_f and AOD_t. In our previous research, although higher-precision results of AOD_f and AOD_t have been obtained, the FMF error is related to the error of the two retrieval parameters. The transmission of the error will eventually amplify the retrieval error of FMF. Compared with the individual retrieval of AOD_f and AOD_t, the retrieval of FMF is still difficult. In the future, it is still necessary to further improve the retrieval accuracy of AOD_f and AOD_t. to obtain more accurate FMF results. In this way, some applications that rely on FMF (such as using the PMRS model to estimate $PM_{2.5}$ concentration) can have better performance. In addition, due to the limitation of the validation data, we are temporarily unable to further discuss the correctness of the spatial distribution trend of the FMF in this study and GRASP, and only the results of the ratio of $PM_{2.5}$ to PM_{10} were used for indirect comparison. In the future, we can try to perform FMF retrieval in other regions with many ground-based observations around the world to further compare the findings of the two results.

Data availability. The FMF datasets produced in this study can be requested from the corresponding author(lizq@radi.ac.cn).

Author contributions. ZL conceived and designed the study. YZ and LQ collected and processed the remote sensing data. YZ and YW performed the FMF retrievals. YZ and YX compared the retrieval results with the AERONET, MODIS, GRASP products. WH and LL ~~analyzed~~ analysed the spatiotemporal trends of FMF in China. ZL and YW collected and processed the in situ data. YZ and ZL prepared the paper with contributions from all coauthors.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. This work was supported by the National Natural Science Fund of China (41901294, 41771535), the National Natural Science Foundation of Chongqing, China (cstc2019jcyj-msxm0726), the Scientific Research Foundation of CUIT (KYTZ201909), the Science and Technology Department of Sichuan Province Foundation (2019YFS0470), and the Chengdu Science and technology project (2018-ZM01-00037-SN).

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Table **1. AERONET site information employed in this study. The land cover types are from the MODIS MCD12 landcover product.**

AERONET sites	Longitude (°E)	Latitude (°N)	Land cover type
Beijing	116.381	39.977	Urban
Hangzhou_City	120.157	30.290	Urban
Hefei	117.162	31.905	Urban
Hong_Kong_PolyU	114.180	22.303	Urban
Kaiping	112.539	22.315	Urban
Lanzhou_City	103.853	36.048	Urban
Minqin	102.959	38.607	Barren
NAM_CO	90.962	30.773	Grasslands
NUIST	118.717	32.206	Urban
QOMS_CAS	86.948	28.365	Barren
SACOL	104.137	35.946	Grasslands
Taihu	120.215	31.421	Wetlands
Taipei_CWB	121.538	25.015	Urban
Xianghe	116.962	39.754	Croplands
Xinglong	117.578	40.396	Forests
Zhongshan_Univ	113.390	23.060	Urban

Table **2. FMF validation results of different surface types**

Land cover type	N	r	MAE	RMSE	Within EE
Overall result	1186	0.770	0.143	0.170	6~~0.96~~5.01%
Urban	421	0.733	0.139	0.163	66.~~03~~98%
Barren	63	0.711	0.158	0.182	~~42~~55.~~86~~55%
Grasslands	113	0.777	0.137	0.170	~~61~~66.~~06~~37%
Wetlands	150	0.508	0.145	0.176	~~62~~63.~~.00~~33%
Croplands	394	0.651	0.146	0.174	~~57~~64.~~86~~21%
Forests	45	0.831	0.133	0.159	~~66~~68.~~66~~88%

Table 3. Statistical analysis of AOD_f and AOD_t ~~errors~~bias

Land cover type	Retrieval parameter (550 nm)	N	r	~~Mean error~~Bias	Proportion of negative ~~offset~~bias	Proportion of positive ~~offset~~bias
Barren	AOD_f ~~(550 nm)~~	63	0.574	-~~0.039~~0.006	~~61.96~~44.44%	55.56~~38.04~~%
	AOD_t ~~(550 nm)~~		0.448	0.111~~0.043~~	1.59~~44.57~~%	98.41~~55.43~~%
	FMF ~~(550 nm)~~		0.711	-0.144~~078~~	~~68.47~~87.30%	12.70~~31.53~~%
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%

Table 4 Comparison between the retrieved and MODIS FMF

Retrieval parameter	MAE (this study)	RMSE (this study)	Within EE (this study)	MAE (MODIS)	RMSE (MODIS)	Within EE (MODIS)
FMF (550 nm)	0.072	0.102	~~79.72~~87.41 %	0.512	0.574	~~12~~19.~~59~~58%

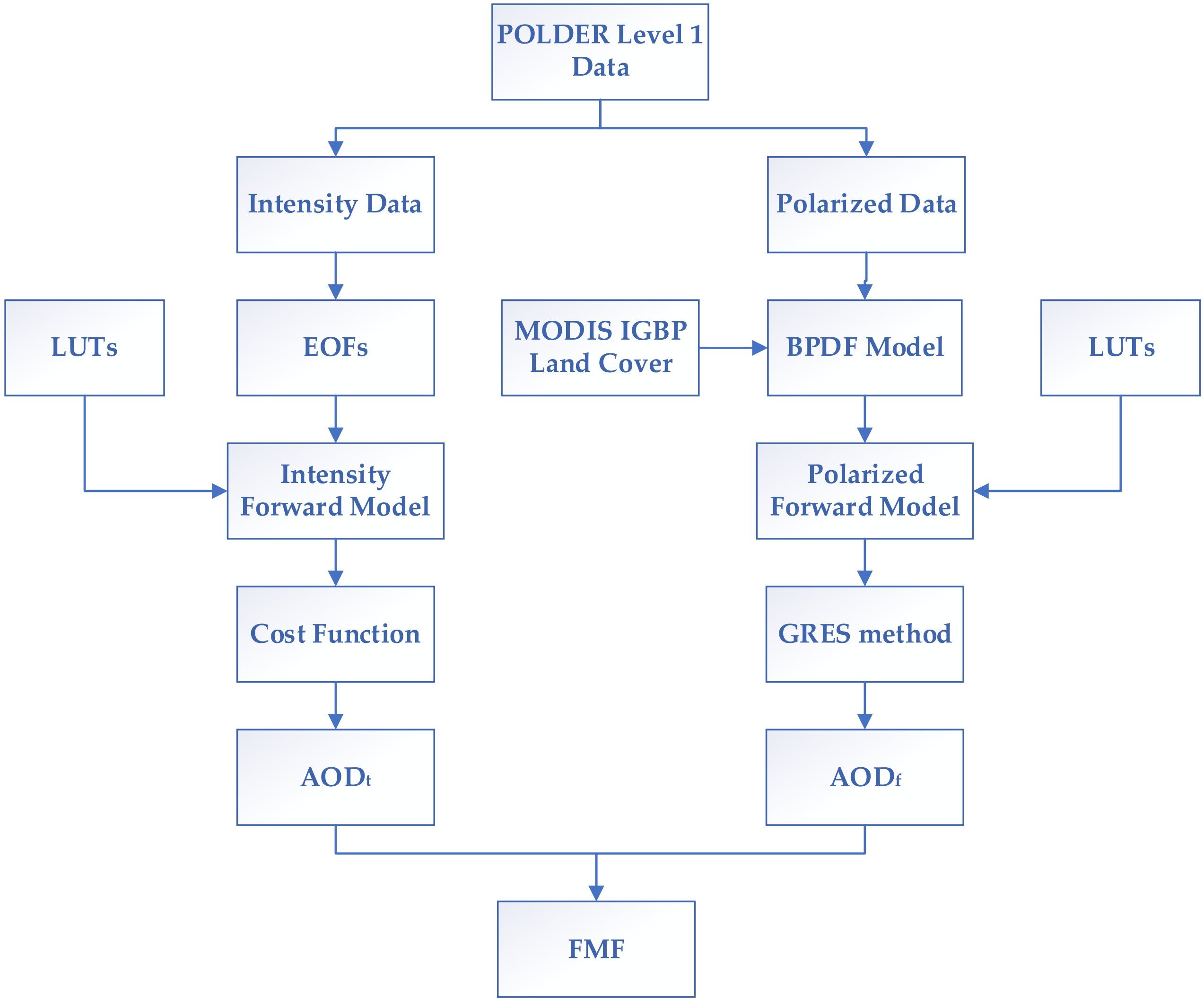


Figure 1. FMF retrieval technology framework of this research

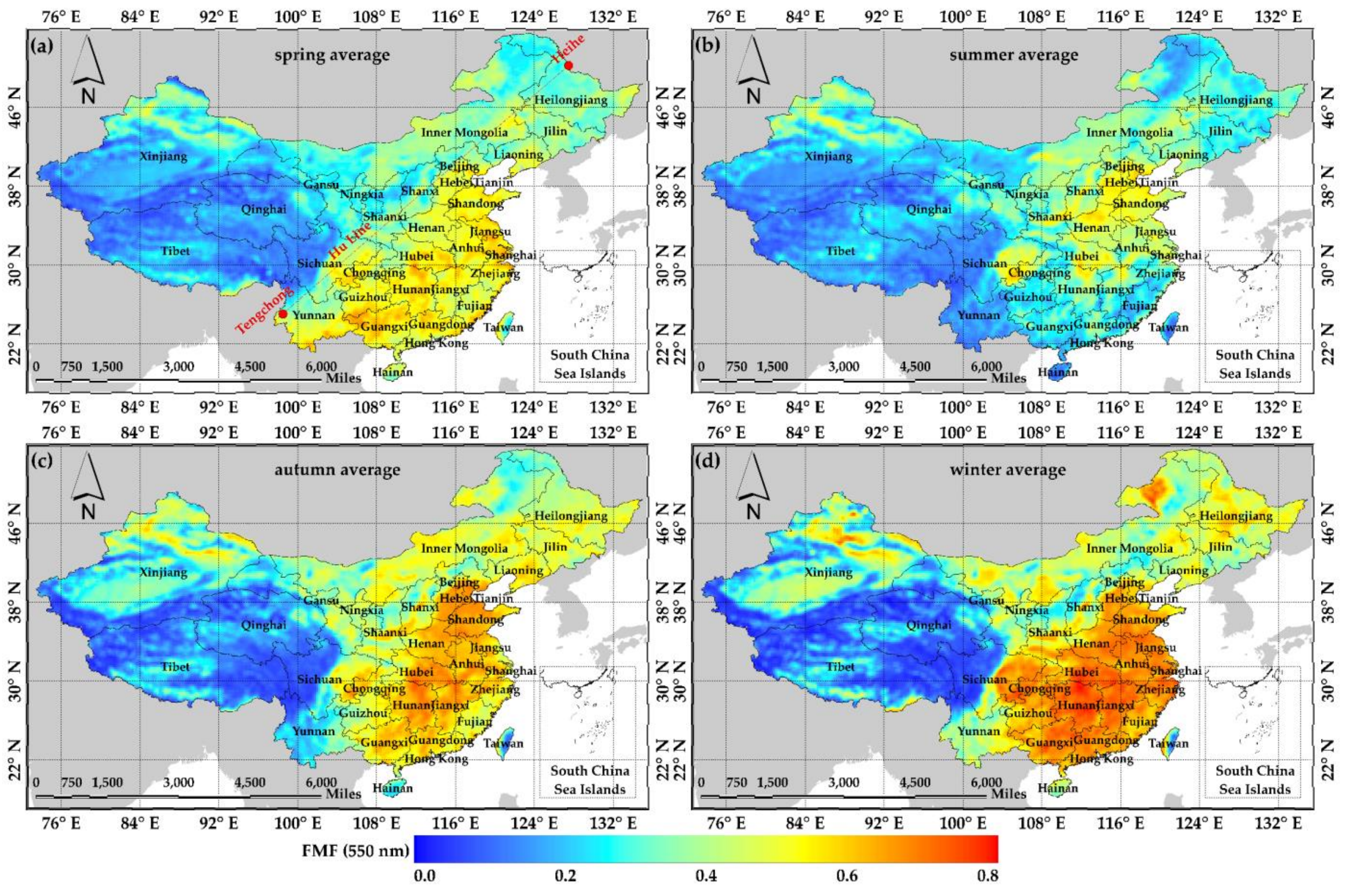


Figure ~~17~~2. Results of the FMF seasonal average spatial distribution of China. (a)-(d) are the results of spring, summer, autumn and winter, respectively, from 2006 to 2013.

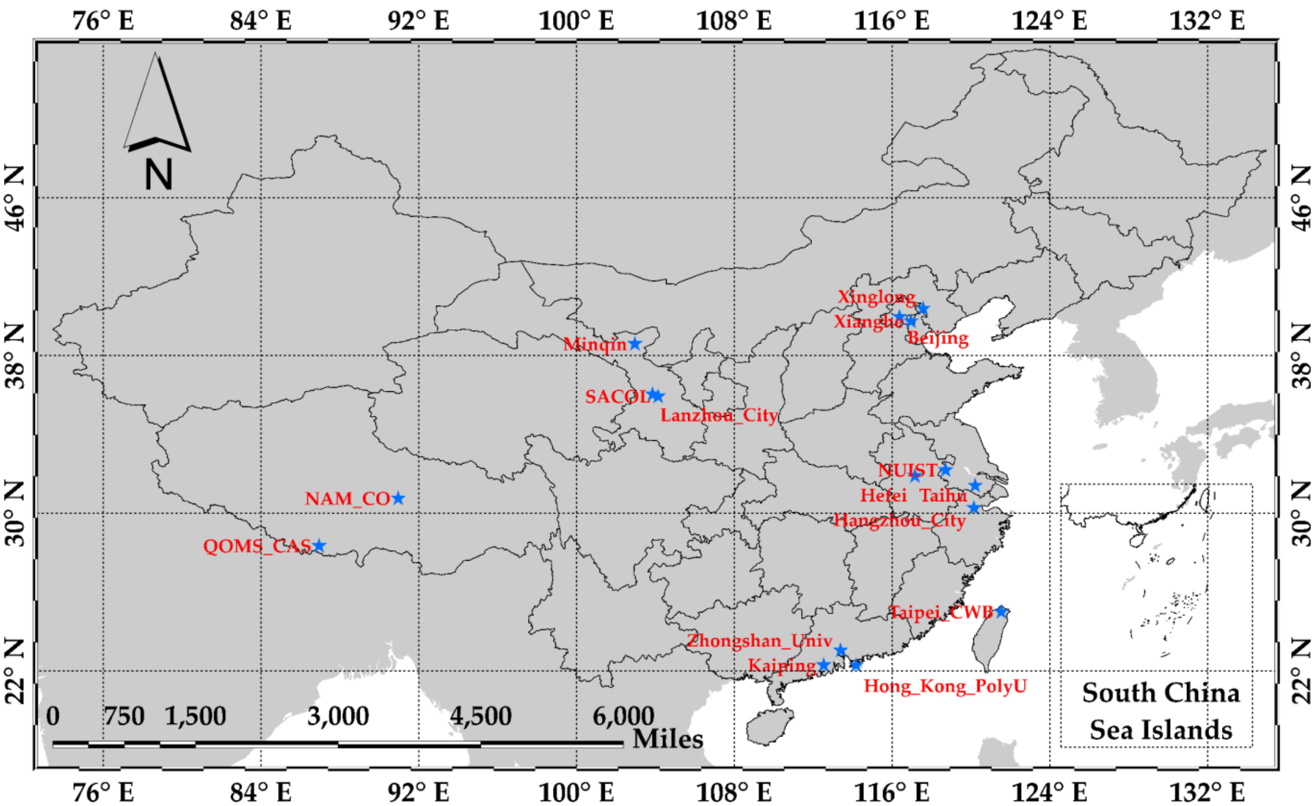
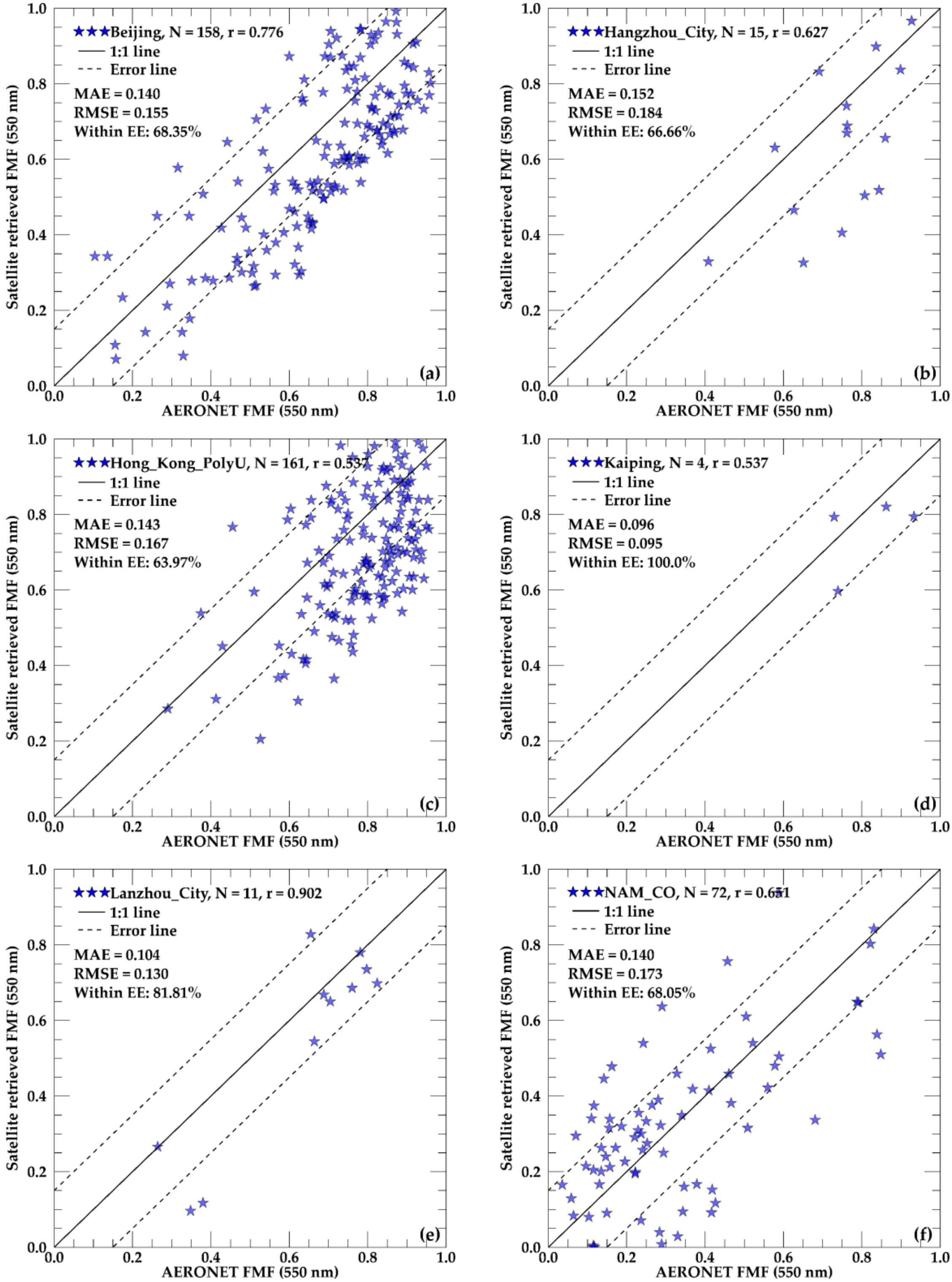
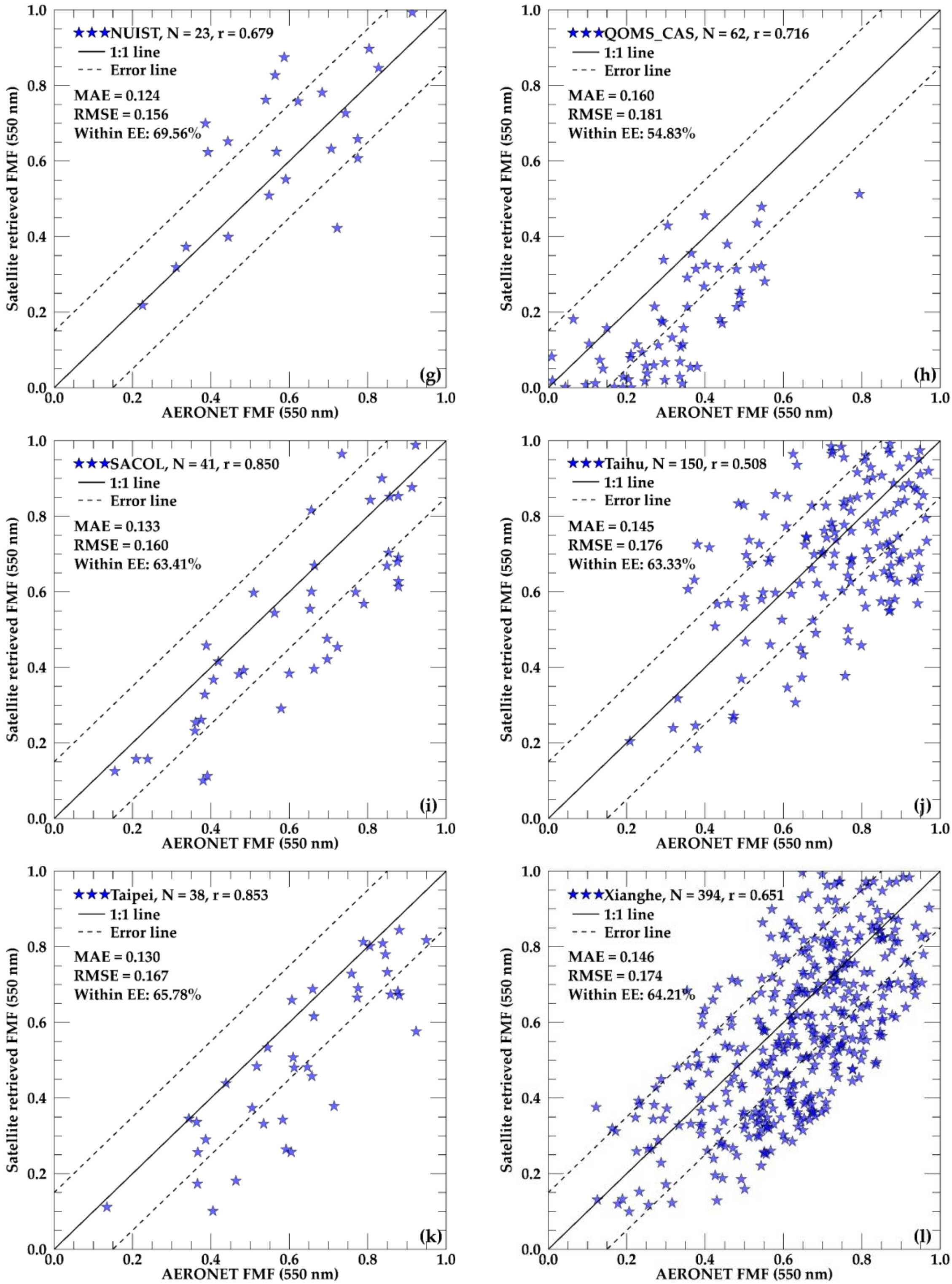


Figure ~~2~~3. The spatial distribution of AERONET sites selected in this study


Beijing, N = 158, r = 0.776
1:1 line
Error line
MAE = 0.140
RMSE = 0.155
Within EE: 68.35%
(a)
Hangzhou_City, N = 15, r = 0.627
1:1 line
Error line
MAE = 0.152
RMSE = 0.184
Within EE: 66.66%
(b)
Hong_Kong_PolyU, N = 161, r = 0.537
1:1 line
Error line
MAE = 0.143
RMSE = 0.167
Within EE: 63.97%
(c)
Kaiping, N = 4, r = 0.537
1:1 line
Error line
MAE = 0.096
RMSE = 0.095
Within EE: 100.0%
(d)
Lanzhou_City, N = 11, r = 0.902
1:1 line
Error line
MAE = 0.104
RMSE = 0.130
Within EE: 81.81%
(e)
NAM_CO, N = 72, r = 0.651
1:1 line
Error line
MAE = 0.140
RMSE = 0.173
Within EE: 68.05%
(f)
Satellite retrieved FMF (550 nm)
AERONET FMF (550 nm)
0.0
0.2
0.4
0.6
0.8
1.0


NUIST, N = 23, r = 0.679
1:1 line
Error line
MAE = 0.124
RMSE = 0.156
Within EE: 69.56%
(g)
QOMS_CAS, N = 62, r = 0.716
1:1 line
Error line
MAE = 0.160
RMSE = 0.181
Within EE: 54.83%
(h)
SACOL, N = 41, r = 0.850
1:1 line
Error line
MAE = 0.133
RMSE = 0.160
Within EE: 63.41%
(i)
Taihu, N = 150, r = 0.508
1:1 line
Error line
MAE = 0.145
RMSE = 0.176
Within EE: 63.33%
(j)
Taipei, N = 38, r = 0.853
1:1 line
Error line
MAE = 0.130
RMSE = 0.167
Within EE: 65.78%
(k)
Xianghe, N = 394, r = 0.651
1:1 line
Error line
MAE = 0.146
RMSE = 0.174
Within EE: 64.21%
(l)
Satellite retrieved FMF (550 nm)
AERONET FMF (550 nm)

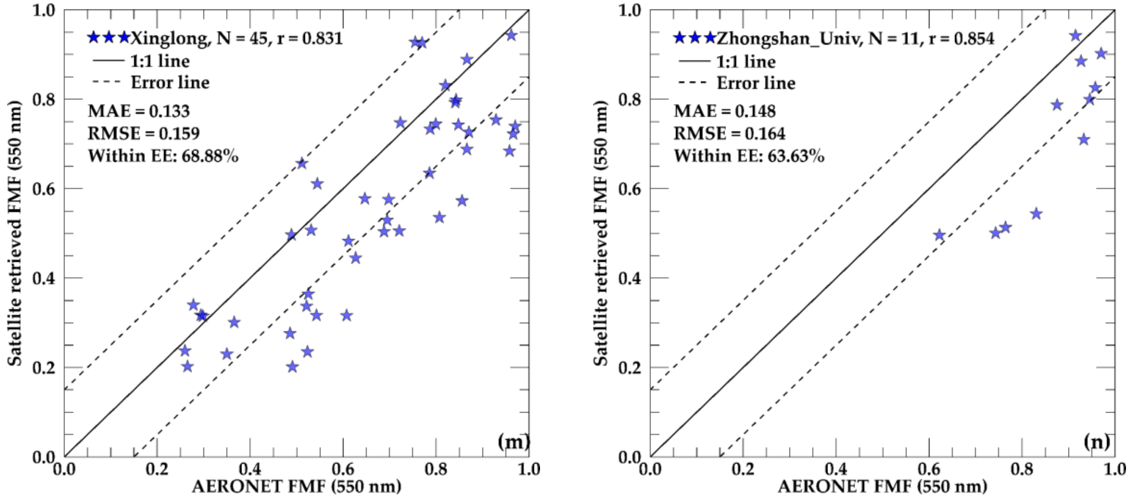


Figure ~~3~~4. FMF results comparison at 14 AERONET sites. (a) - (~~d~~n) are the validation results for the Beijing, Hangzhou_city, Hongkong_PolyU, Kaiping, Lanzhou_city, NAM_CO, NUIST, QOMS_CAS, SACOL, Taihu, Taipei, Xianghe, Xinglong, Zhongshan_Univ sites, respectively.

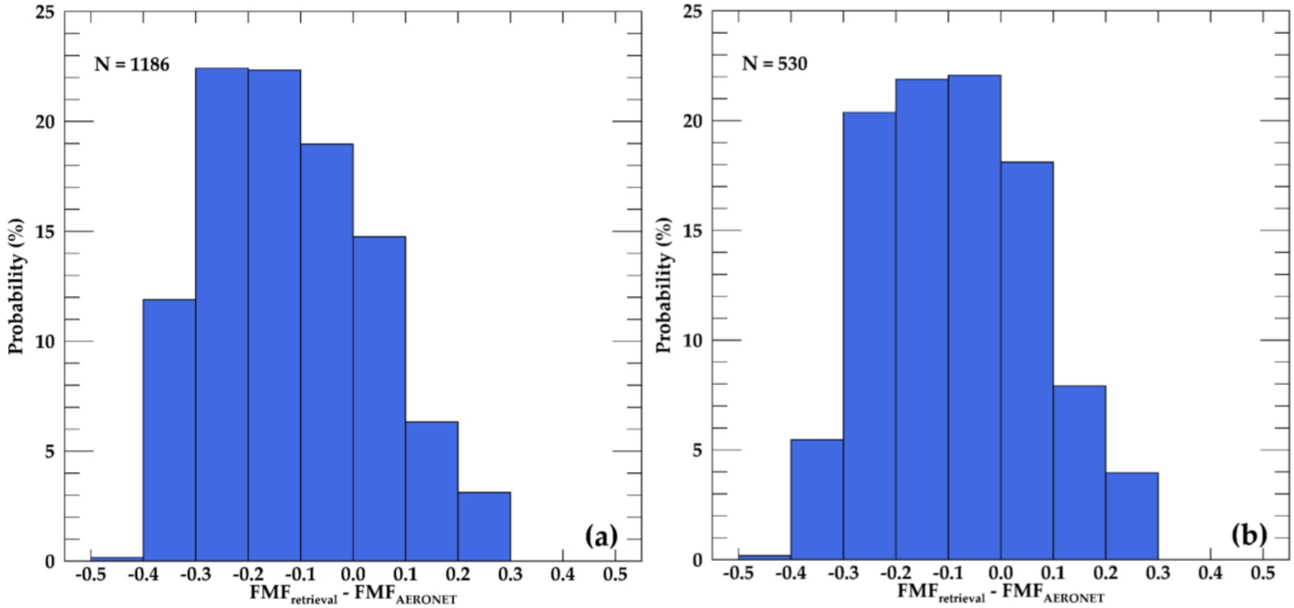
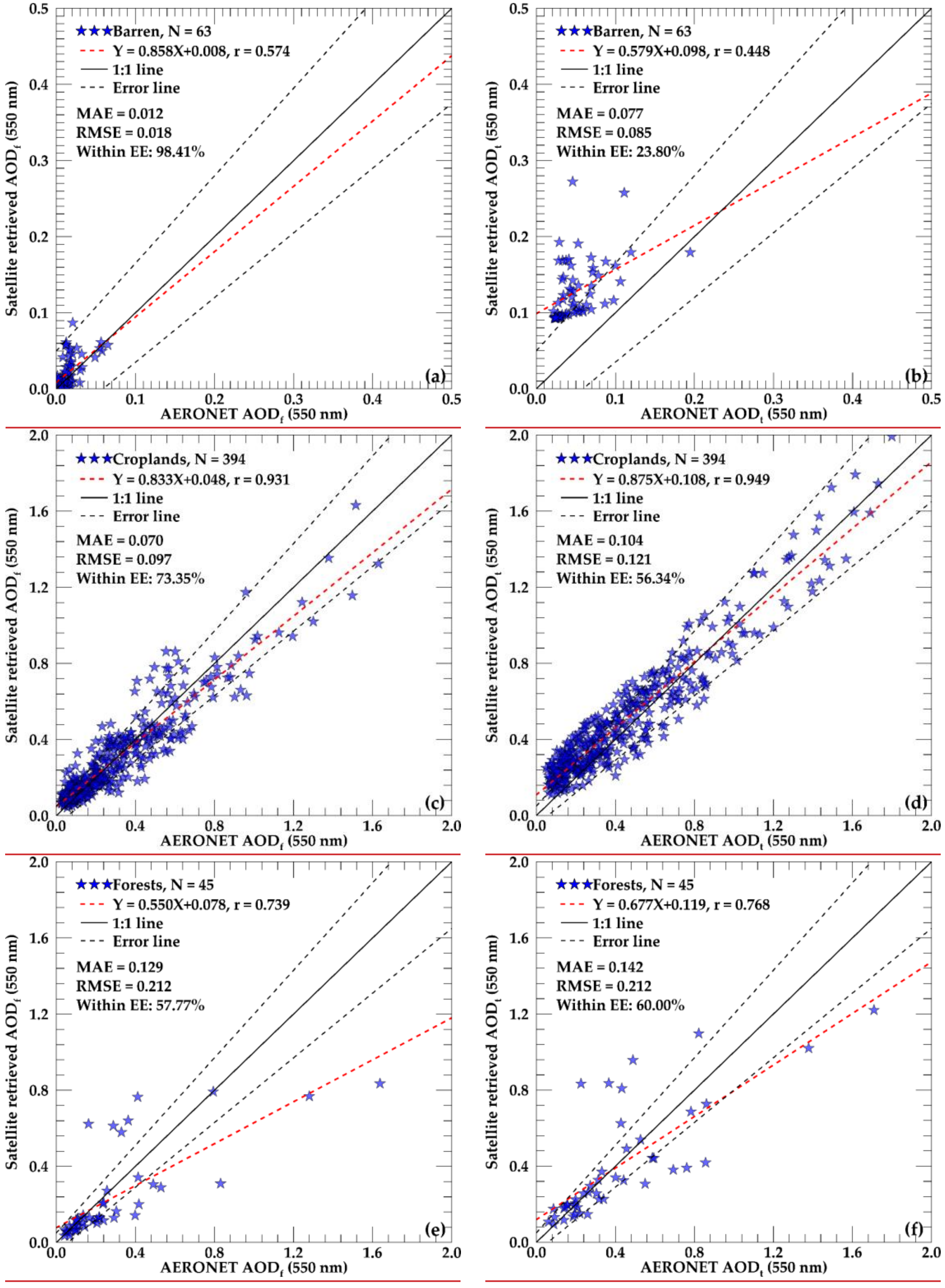


Figure 45. FMF retrieval error distribution results. (a) is for all results, and (b) is for the results with AODf greater than 0.2.


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)
AERONET AOD$_f$ (550 nm)
Satellite retrieved 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)
AERONET AOD$_t$ (550 nm)
Satellite retrieved 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)
AERONET AOD$_f$ (550 nm)
Satellite retrieved 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)
AERONET AOD$_t$ (550 nm)
Satellite retrieved 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)
AERONET AOD$_f$ (550 nm)
Satellite retrieved 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)
AERONET AOD$_t$ (550 nm)
Satellite retrieved AOD$_t$ (550 nm)

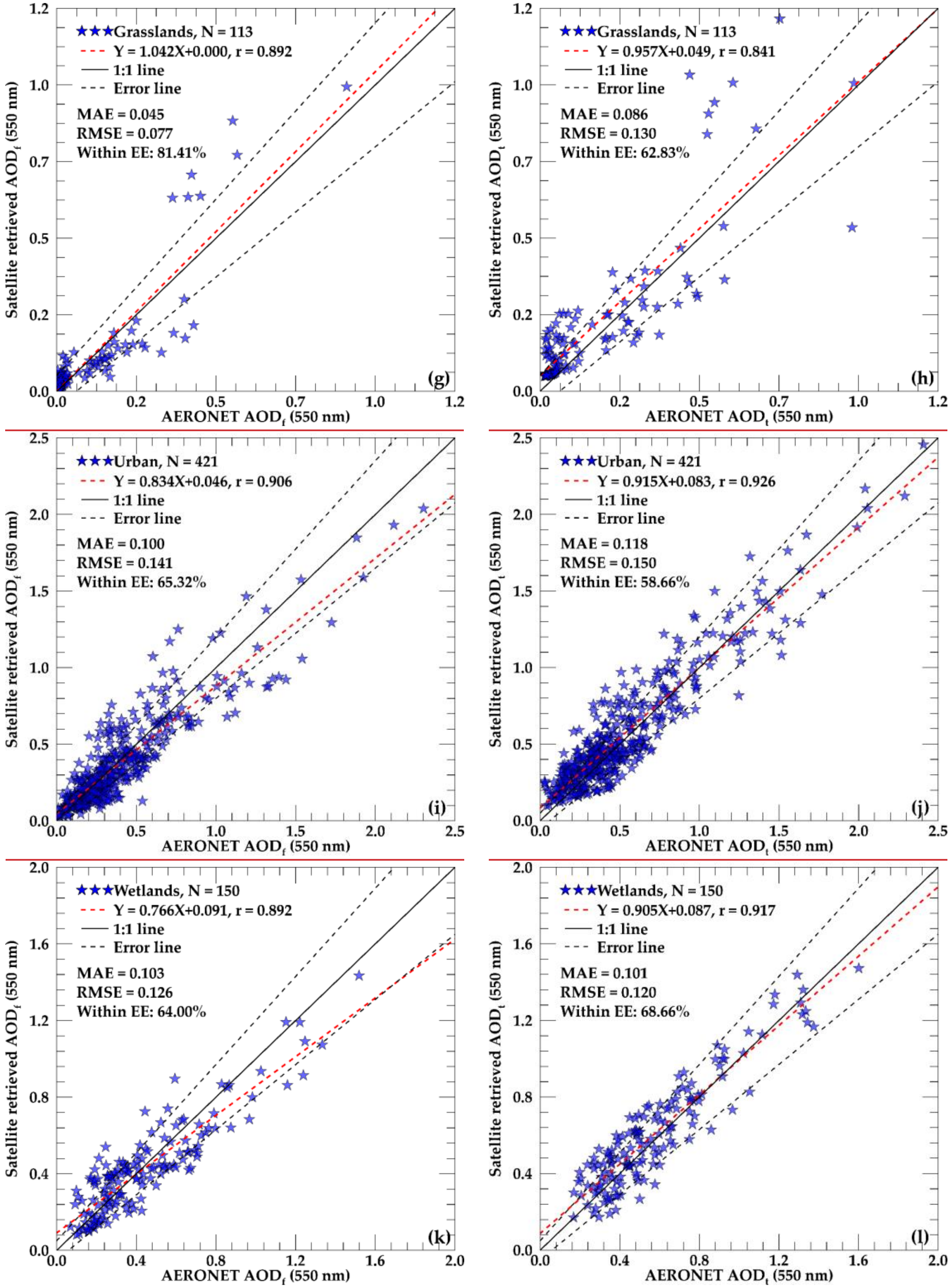

Grasslands, N = 113
Y = 1.042X+0.000, r = 0.892
1:1 line
Error line
MAE = 0.045
RMSE = 0.077
Within EE: 81.41%
(g)
AERONET AOD_f (550 nm)
Satellite retrieved AOD_f (550 nm)
Grasslands, N = 113
Y = 0.957X+0.049, r = 0.841
1:1 line
Error line
MAE = 0.086
RMSE = 0.130
Within EE: 62.83%
(h)
AERONET AOD_t (550 nm)
Satellite retrieved AOD_t (550 nm)
Urban, N = 421
Y = 0.834X+0.046, r = 0.906
1:1 line
Error line
MAE = 0.100
RMSE = 0.141
Within EE: 65.32%
(i)
AERONET AOD_f (550 nm)
Satellite retrieved AOD_f (550 nm)
Urban, N = 421
Y = 0.915X+0.083, r = 0.926
1:1 line
Error line
MAE = 0.118
RMSE = 0.150
Within EE: 58.66%
(j)
AERONET AOD_t (550 nm)
Satellite retrieved AOD_t (550 nm)
Wetlands, N = 150
Y = 0.766X+0.091, r = 0.892
1:1 line
Error line
MAE = 0.103
RMSE = 0.126
Within EE: 64.00%
(k)
AERONET AOD_f (550 nm)
Satellite retrieved AOD_f (550 nm)
Wetlands, N = 150
Y = 0.905X+0.087, r = 0.917
1:1 line
Error line
MAE = 0.101
RMSE = 0.120
Within EE: 68.66%
(l)
AERONET AOD_t (550 nm)
Satellite retrieved AOD_t (550 nm)

Figure 6. 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.

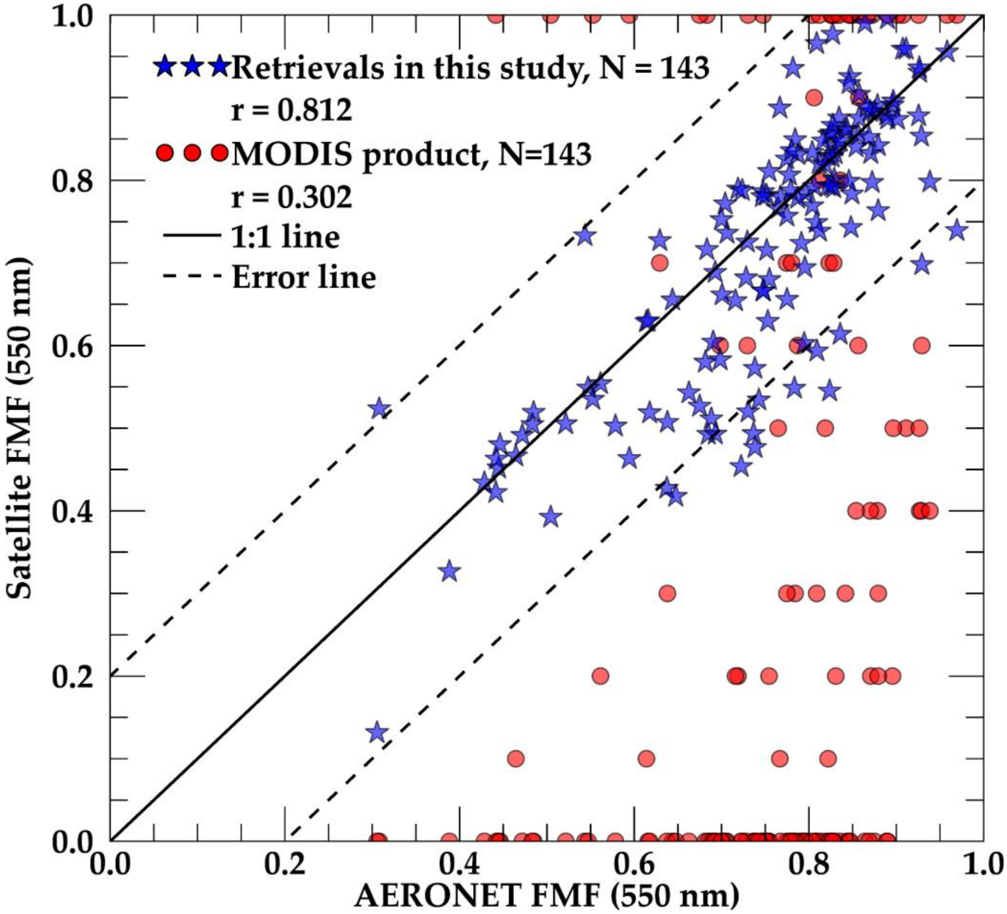


Figure ~~5~~7. Comparison between the results of this study and MODIS FMF with AERONET

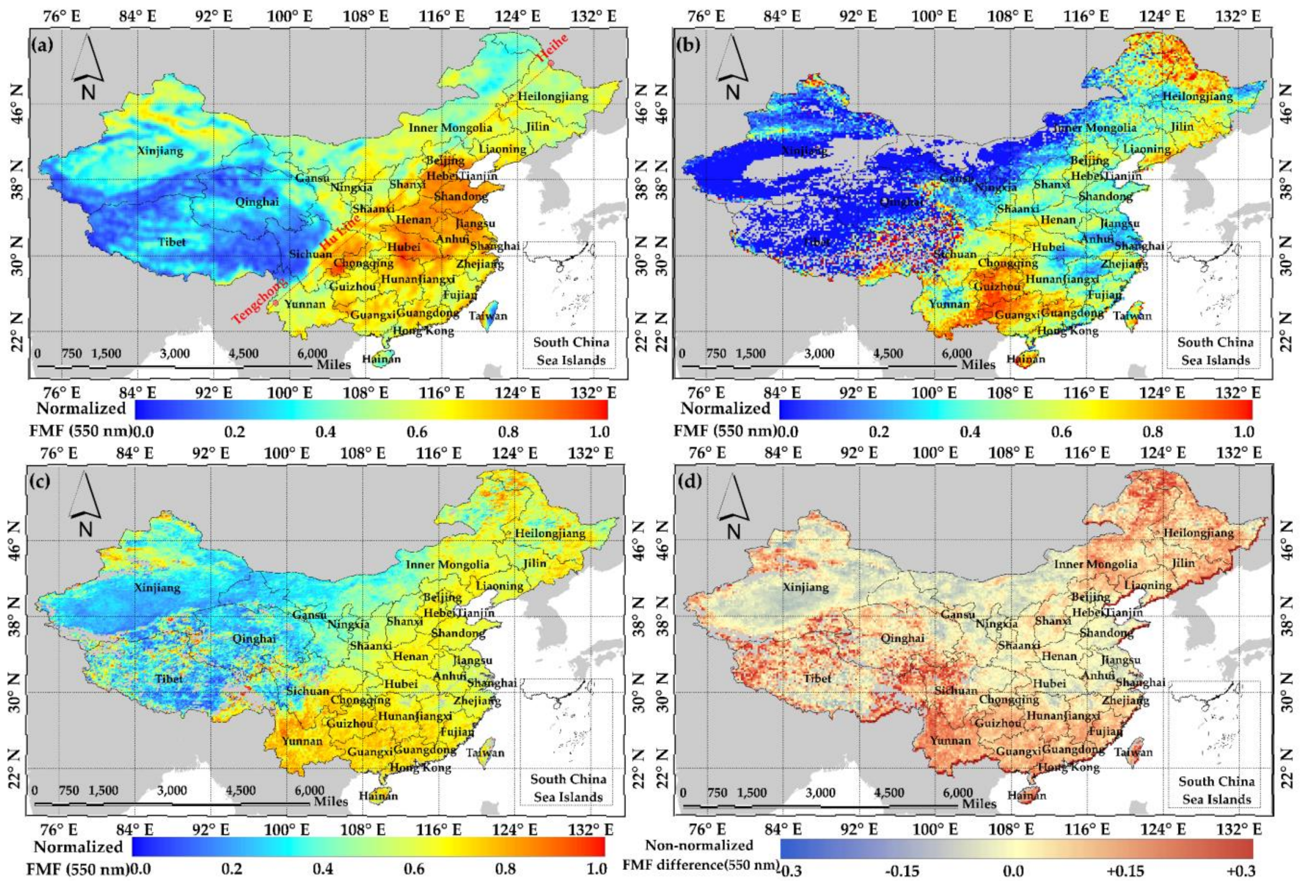
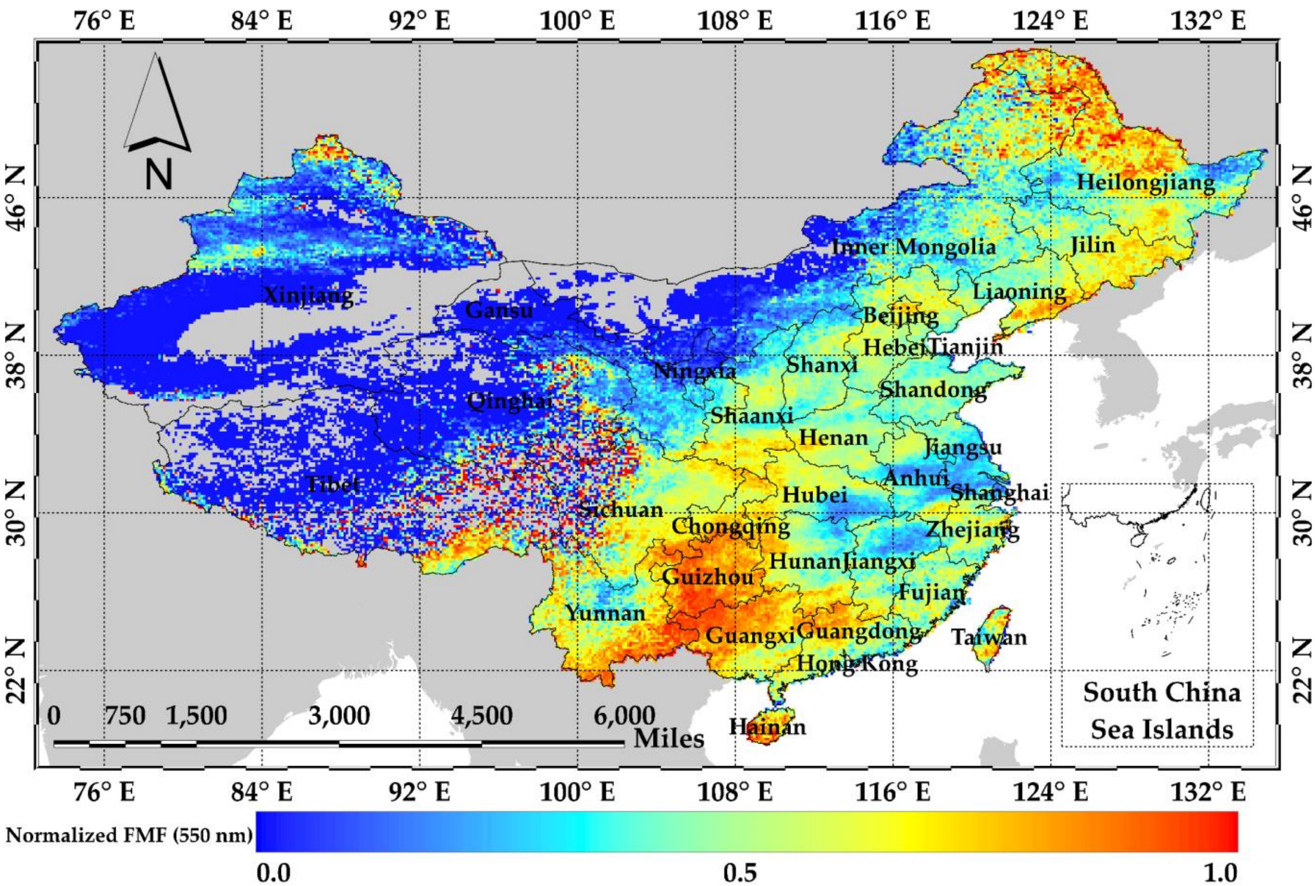
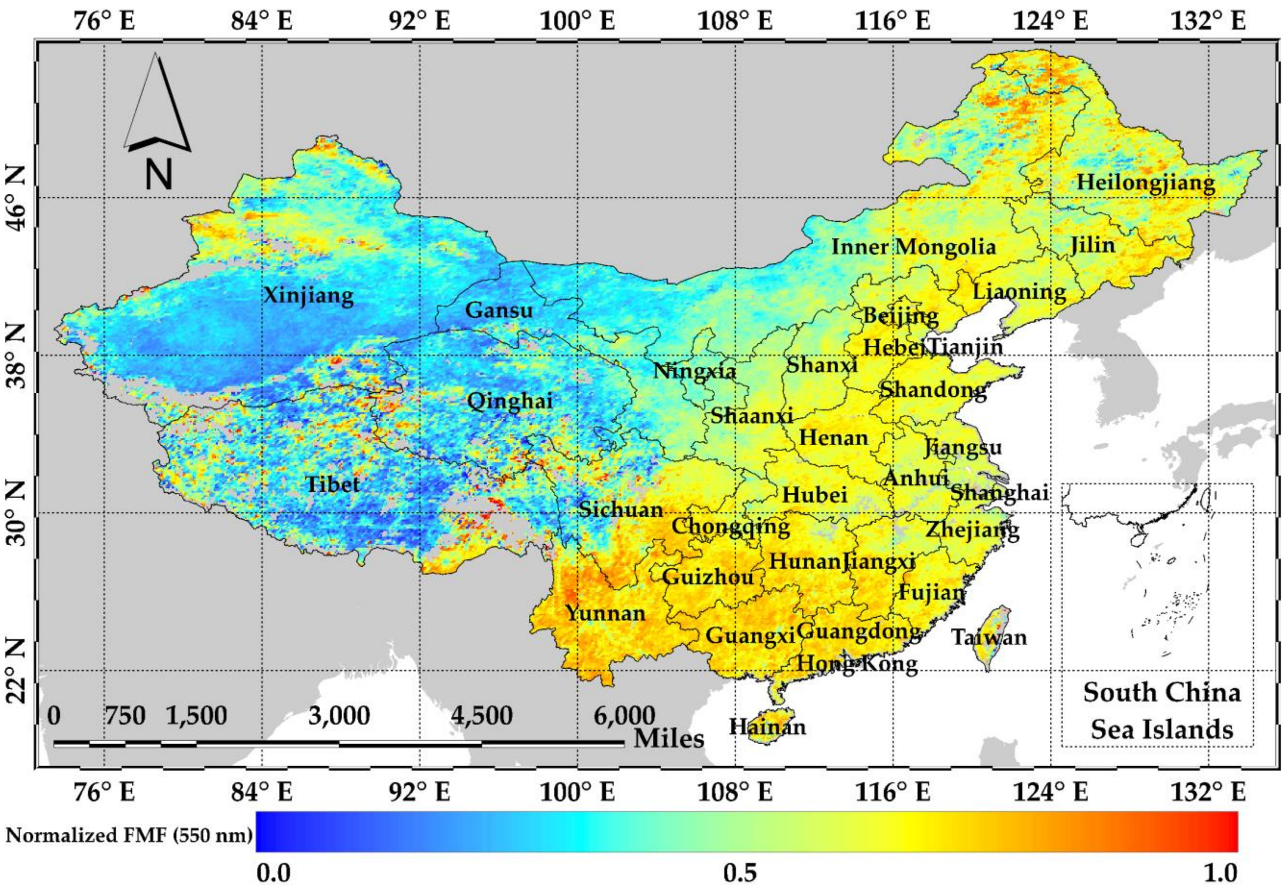


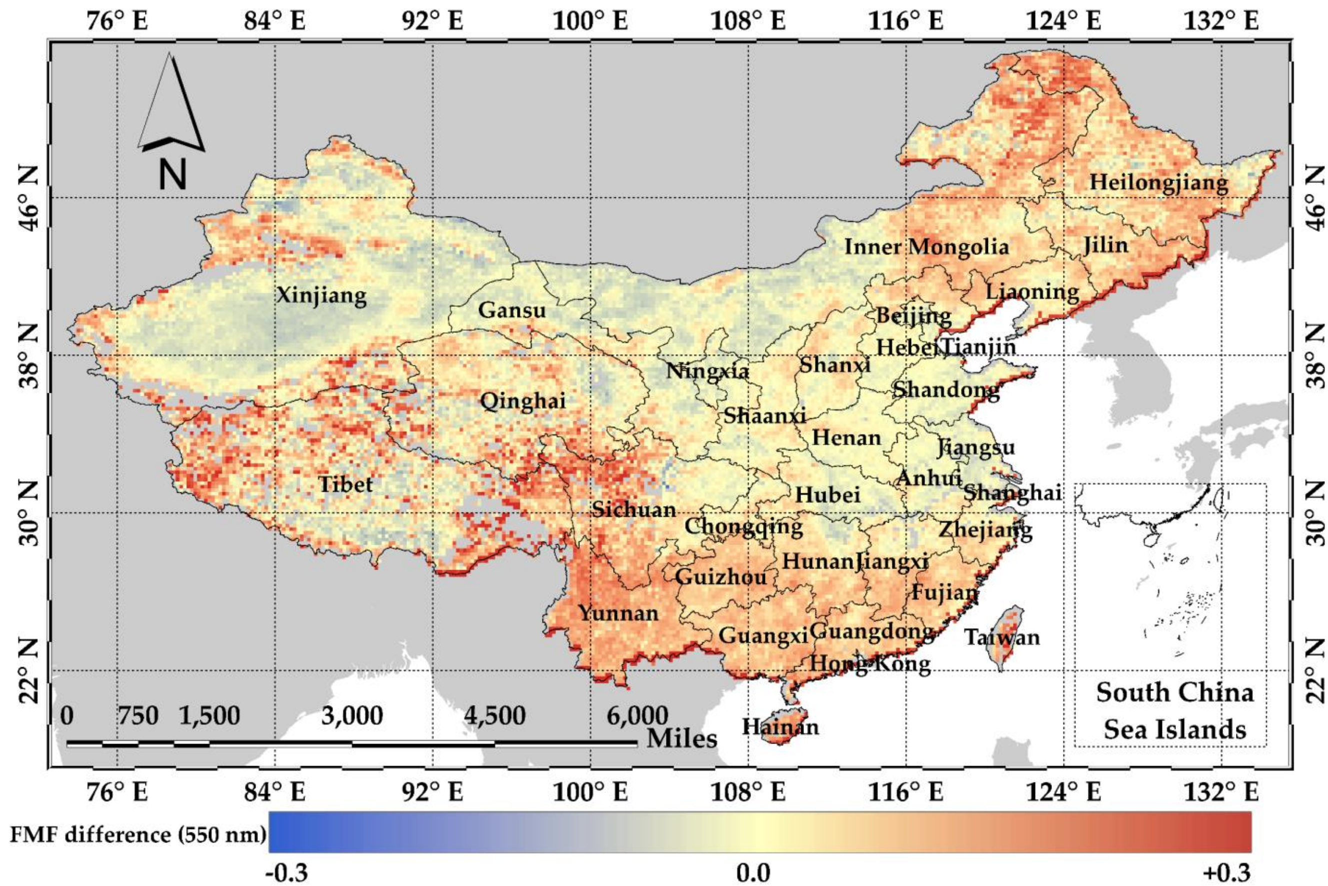
Figure ~~6~~8. Distribution of ~~normalized~~ FMF of China in 2013 from different sources~~ of China in 2013~~. (a) is the normalized results of this study (18 km resolution)~~)~~, (b) is the normalized results of MODIS (10 km resolution), (c) is the normalized results of GRASP (6 km resolution), and (d) is the GRASP results minus the retrieved results (non-normalized, 18 km resolution).



~~**Figure 7. Distribution of normalized FMF of China in 2013 (MODIS results)**~~



~~Figure 8. Distribution of normalized FMF of China in 2013 (GRASP results)~~



655 ~~Figure 9. Differences in FMF results in China in 2013 (GRASP results minus the retrieved results, non-normalized)~~

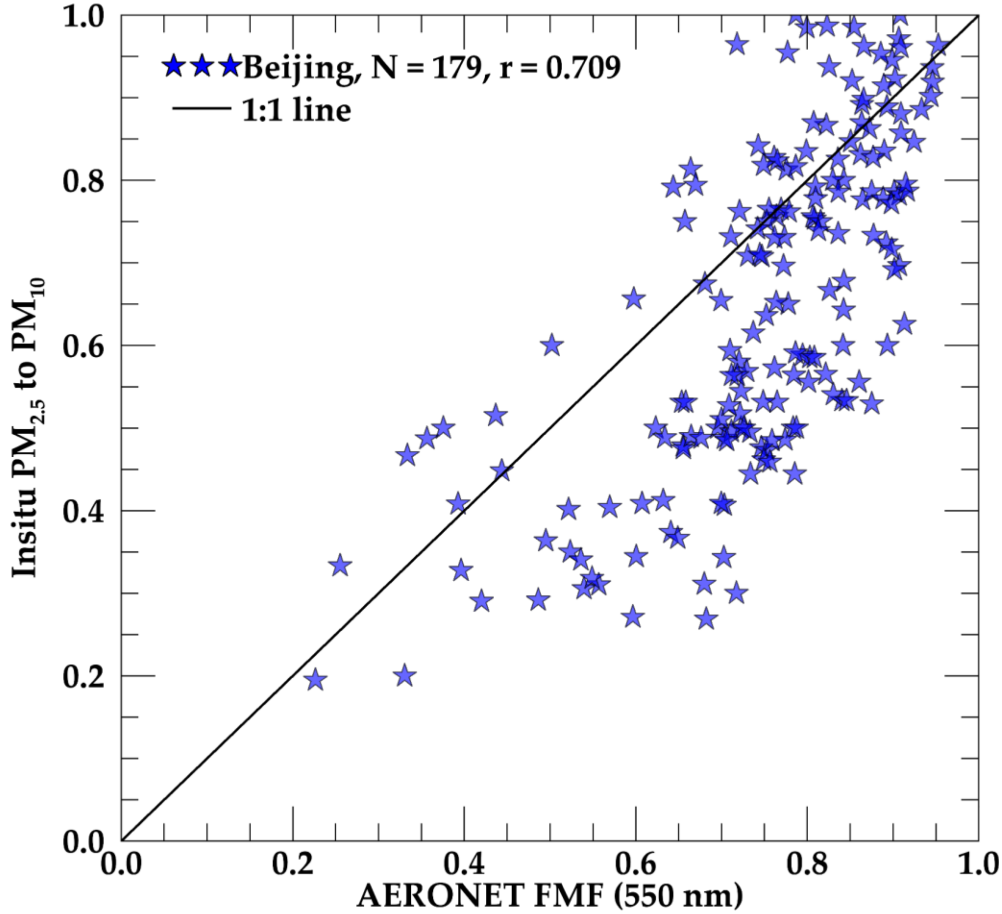


Figure ~~10~~9. Comparison between the ratio of $PM_{2.5}$ to PM_{10} and FMF (hourly average)

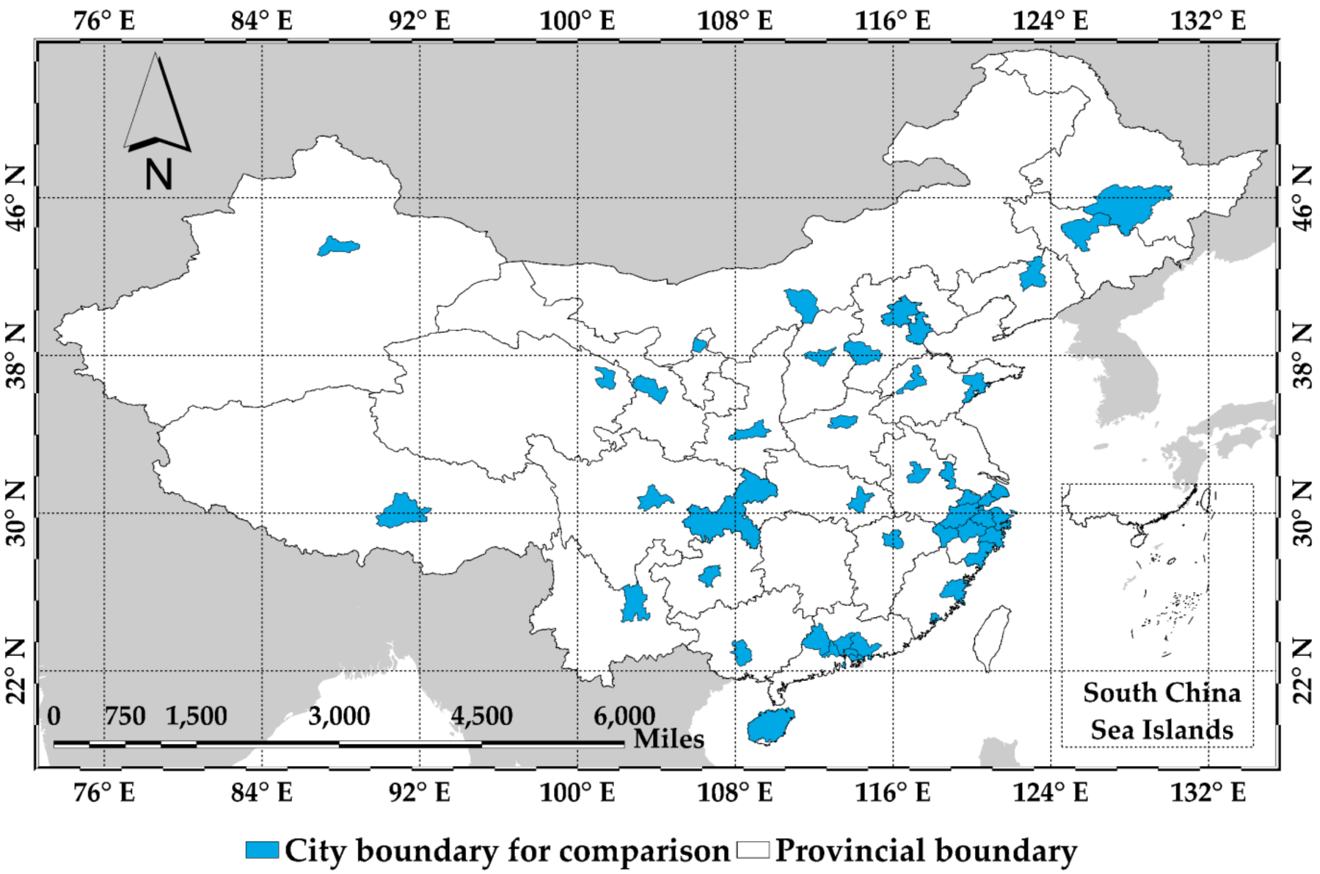


Figure ~~11~~10. Forty-seven urban administrative regions in China used to compare the annual average FMF

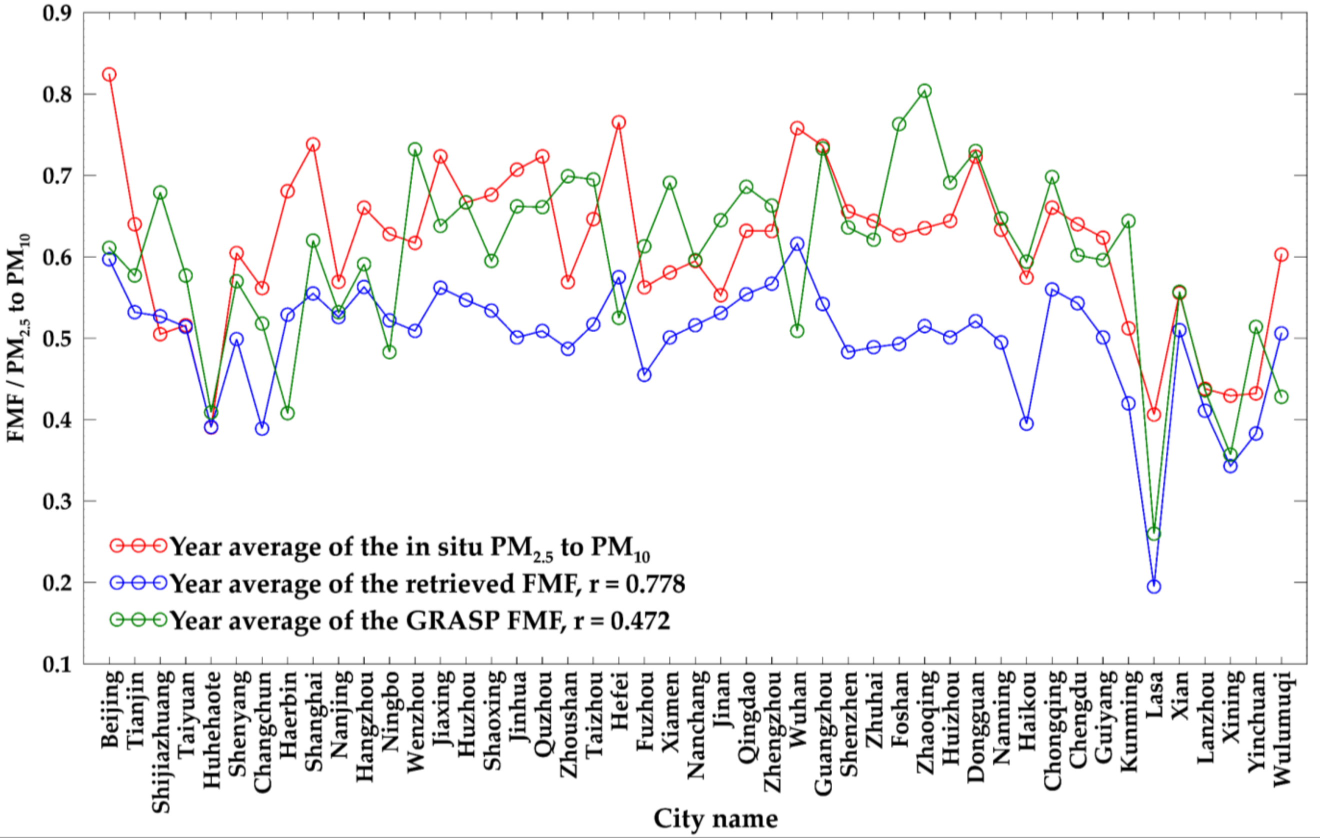
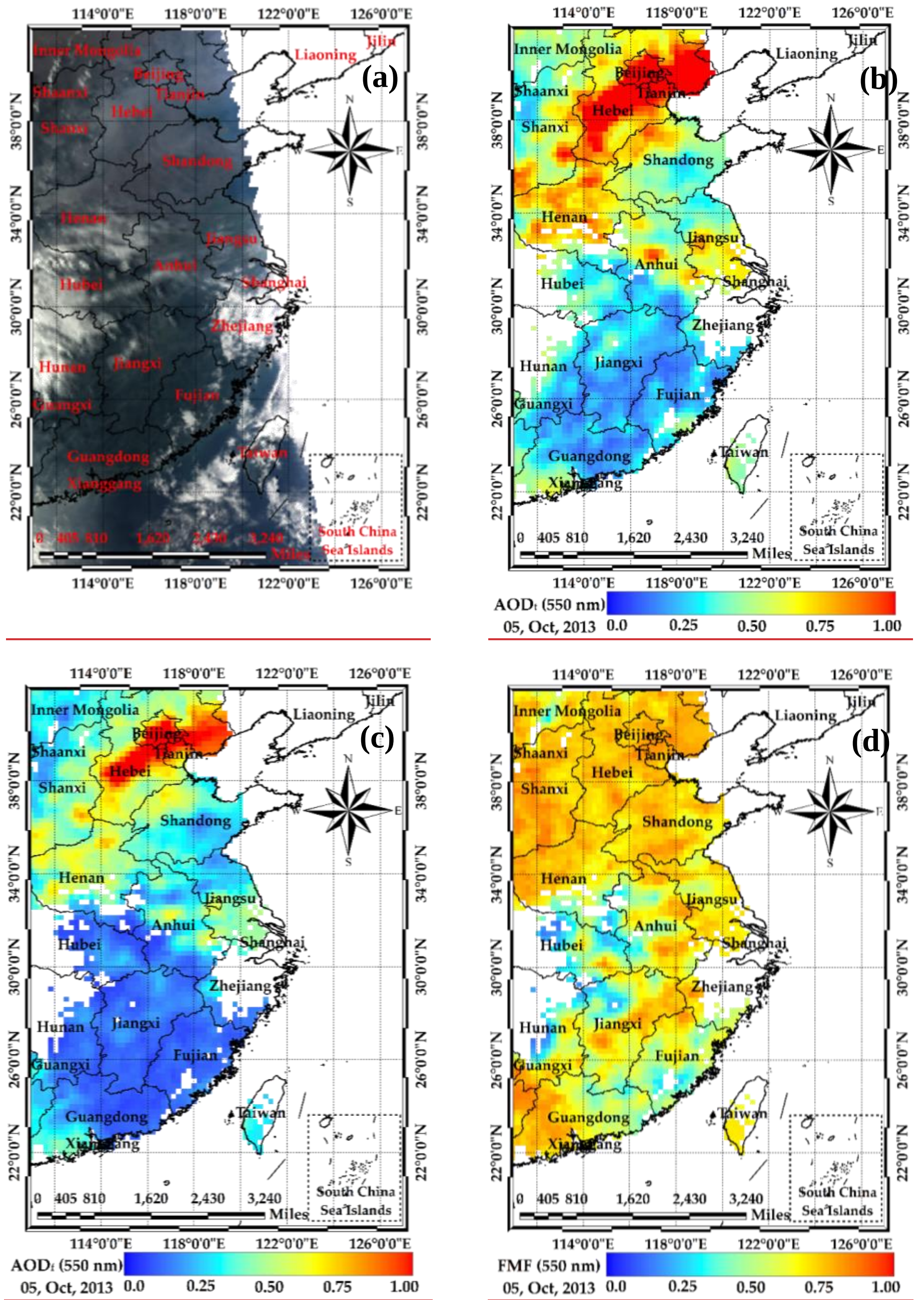
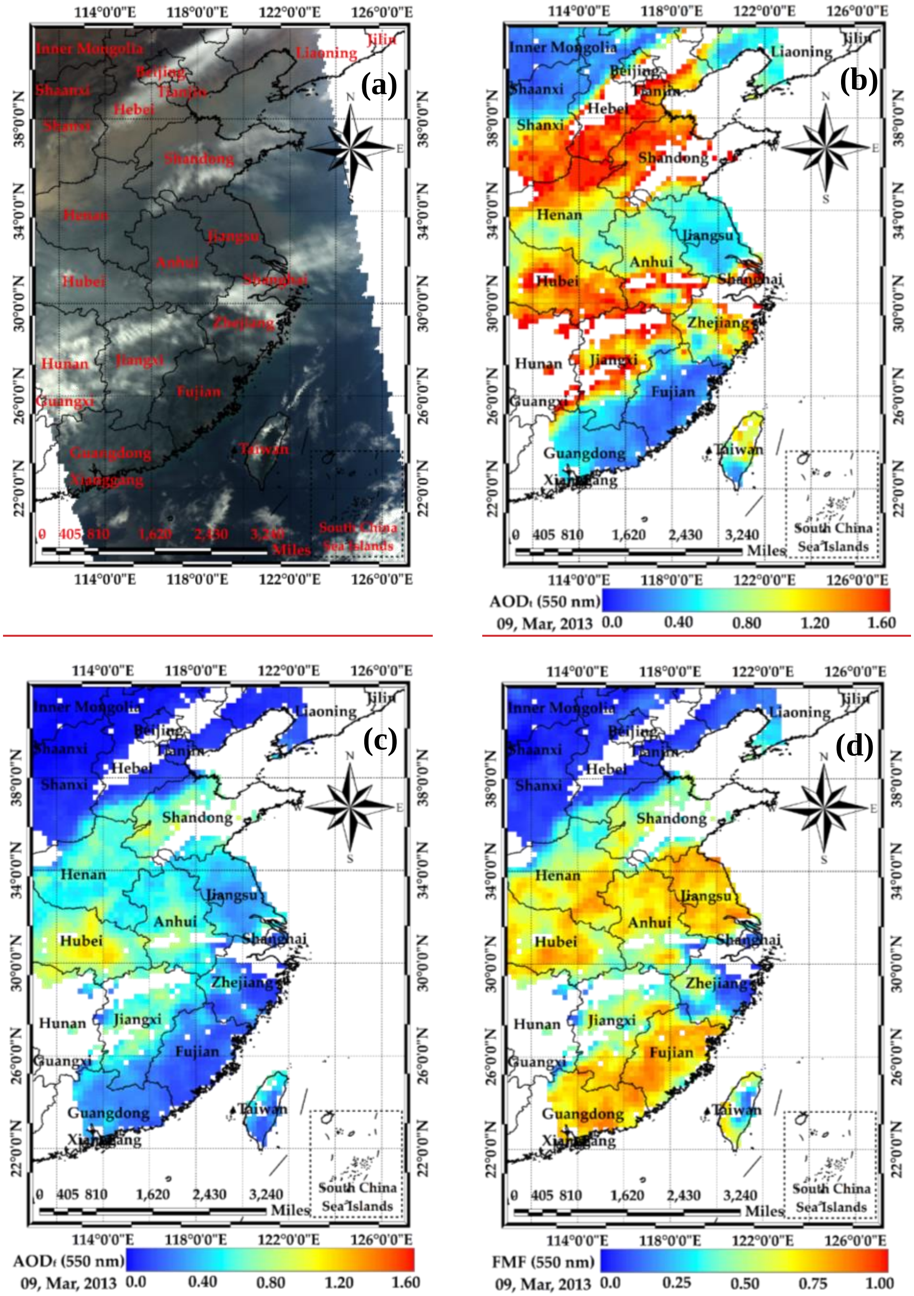


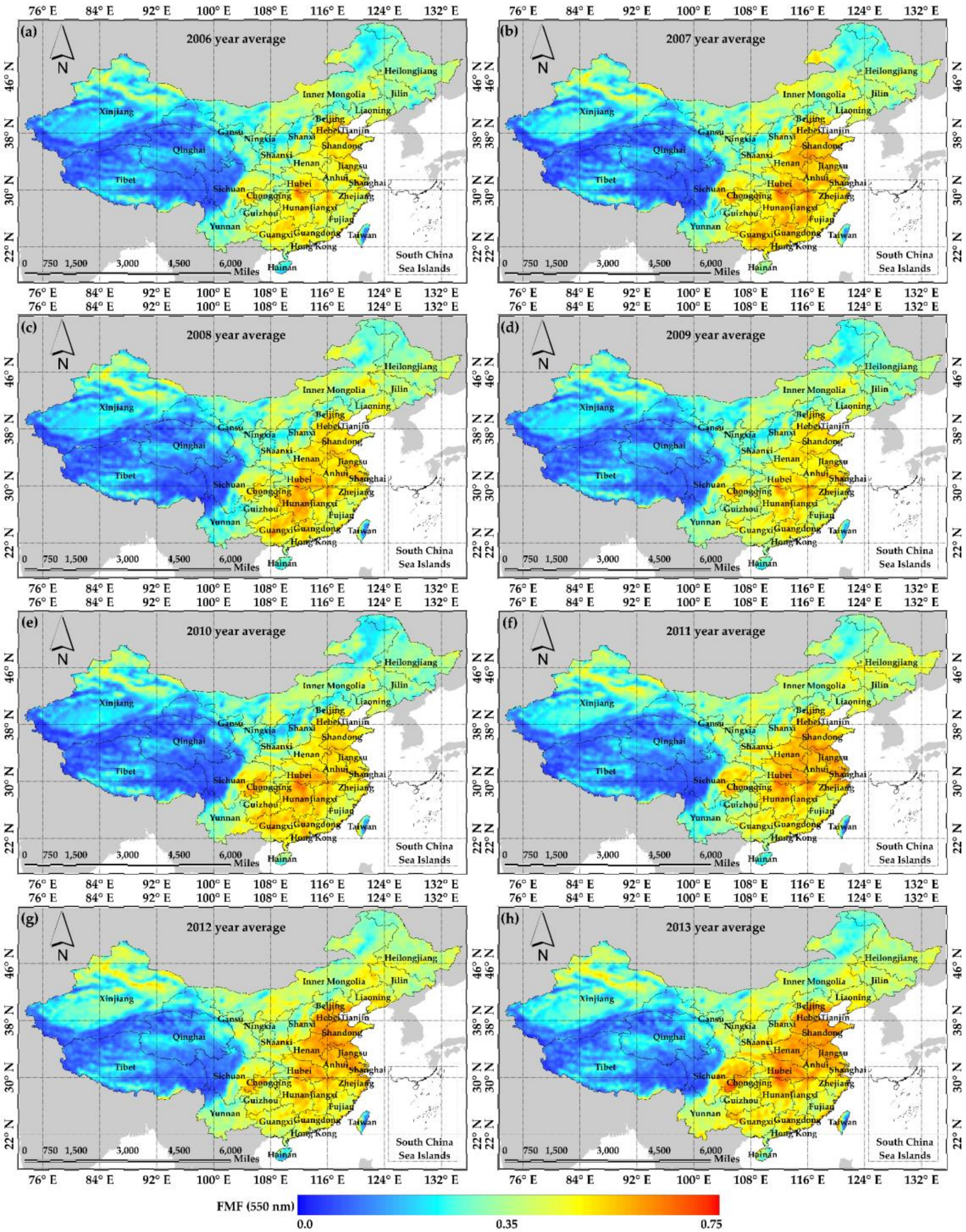
Figure ~~12~~11. Comparison of the results of the retrieved and GRASP FMF with the urban average of the ratio of $PM_{2.5}$ to PM_{10} (2013)



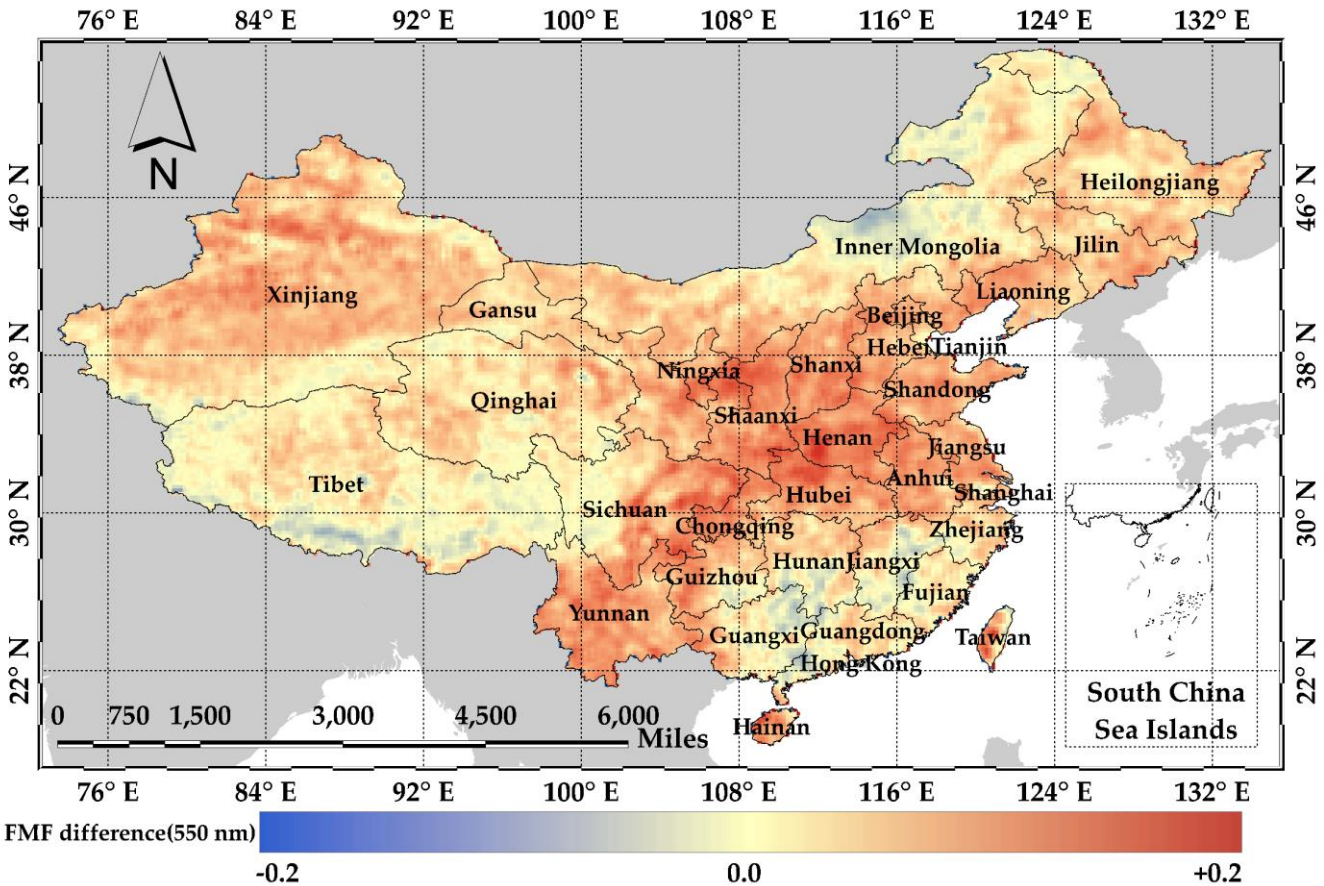
~~**Figure 13. The retrieval result of the haze case in North China on October 5, 2013. (a) is the true colour image of POLDER, (b) is the retrieval result of the AOD_t, (c) is the retrieval result of the AOD_f, and (d) is the retrieval result of the FMF.**~~



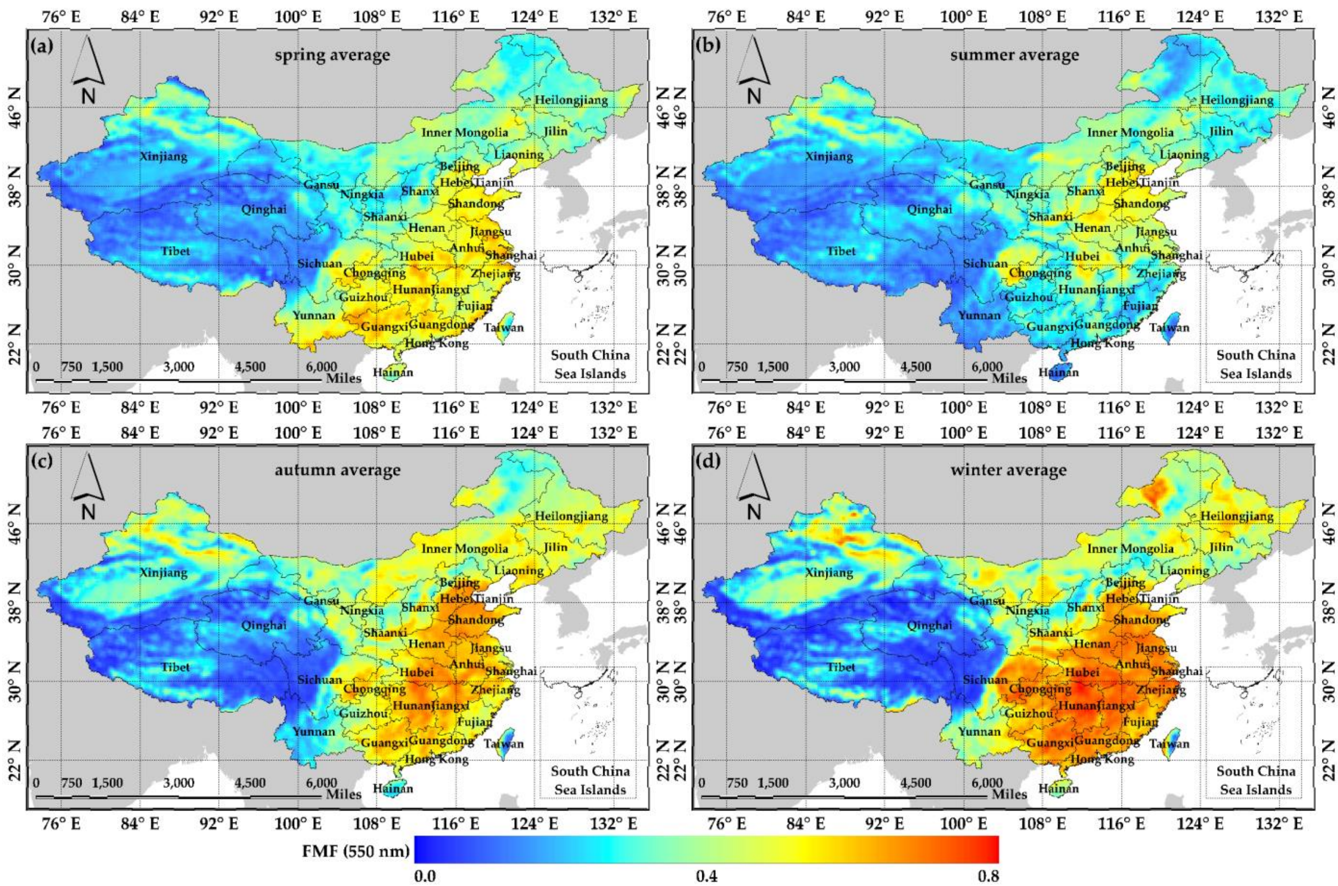
~~**Figure 14. Same as in Figure 13 but for October 5, 2013.**~~



~~**Figure 15. The results of the FMF annual average spatial distribution of China. (a)-(h) are the results from 2006 to 2013, respectively.**~~



~~**Figure 16. Numerical distribution of the spatial variation in FMF in China (2013 minus 2006)**~~



~~**Figure 17. Results of the FMF seasonal average spatial distribution of China. (a)-(d) are the results of spring, summer, autumn and winter from 2006 to 2013.**~~