Responses to RC3:

In this manuscript, the authors developed and presented the new dataset of fine-mode fraction over global land during 2001-2020. The method they proposed is a hybrid physical and deep learning method, which is the physical model output calibration with DL. Generally, this FMF dataset can be useful for studies of anthropogenic aerosol and also the PM2.5 estimation. The paper is written in a consistent workflow and the inter-comparisons in terms of different methods and official products are very comprehensive. However, I think some concerns and issues need to be addressed.

Response: Thank you very much for your constructive suggestions to our manuscript.

Major comments:

1. We know that in deep learning modeling, the test dataset should be independent of the training dataset to avoid the data leakage, therefore my first concern is that the validation for FMF is independent. In Figure 3a, the authors use the AERONET data for training and testing. A more rigorous validation should be added. My suggestion is to conduct independent validation by FMF from other sources of FMF observations, or the Phy-DL FMF is only reliable over the pixels with AERONET sites.

Response: Thank you for this valuable recommendation. We have conducted a validation based on measurements from four independent National Oceanic and Atmospheric Administration Surface Radiation Budget (SURFRAD) network sites not used for training in the deep-learning model. The SURFRAD network provides long-term, multi-band AOD observations at a temporal resolution of three minutes. As shown in **Figure R1**, the four sites (black triangles) are located across the US, covering different land types from forested land to barren land (**Tables R1 and R2**). The land types were based on MODIS MCD12C1 data from the International Geosphere-Biosphere Programme scheme.

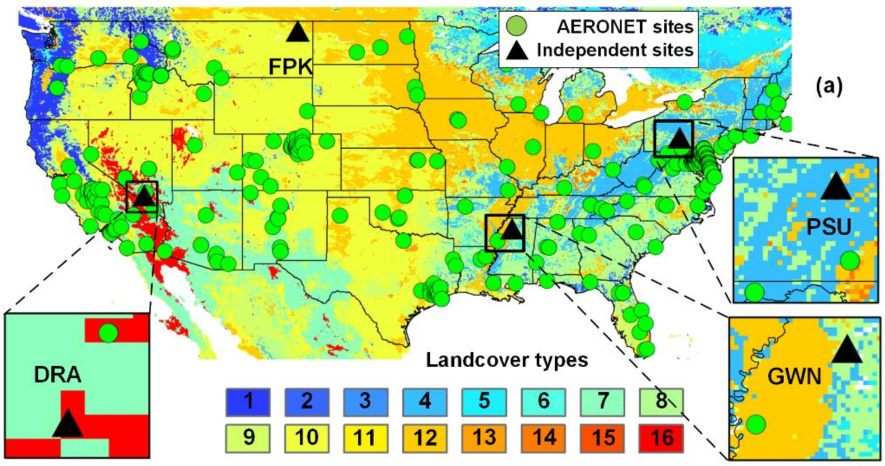

Figure R1. (a) Locations of AERONET sites (green dots) and four independent SURFRAD sites (black triangles, **Table R1**) for the independent validation of the Phy-DL FMF algorithm. The base map shows the land types from MODIS MCD12C1 data (the International Geosphere-Biosphere Programme scheme) in 2010. **Table R2** provides details about the land-type legend.

Table R1. SURFRAD sites used for out-of-site validation and their locations and land types.

Sites	Longitude (ºW)	Latitude (ºN)	Land type
Desert Rock (DRA)	116.02	36.62	Barren or sparse
Fort Peck (FPK)	105.10	48.31	Grasslands
Goodwin Creek (GWN)	89.87	34.25	Woody savannas
Penn State (PSU)	77.93	40.72	Mixed forests

Table R2. Land types and corresponding values from MODIS MCD12C1 data (the International Geosphere-Biosphere Programme scheme).

Value	Land type	Value	Land type
1	Evergreen needleleaf	9	Savannas
2	Evergreen broadleaf	10	Grasslands
3	Deciduous needleleaf	11	Permanent wetlands
4	Deciduous broadleaf	12	Croplands
5	Mixed forests	13	Urban and built up
6	Closed shrubland	14	Crop natural vegetation mosaic
7	Open shrublands	15	Snow and ice
8	Woody savannas	16	Barren or sparse

We used the same AERONET method (SDA) to calculate the FMF at the SURFRAD sites. SURFRAD data were not included in the model training, so SURFRAD FMFs can be regarded as the out-of-site validation for the Phy-DL FMF algorithm. **Figure R2** shows how SURFRAD and Phy-DL FMFs compare. The correlation coefficient (R) was 0.51, and the root-mean-square error (RMSE) was 0.144, somewhat poorer performance than AERONET validation results (i.e., R=0.68 and RMSE=0.136).

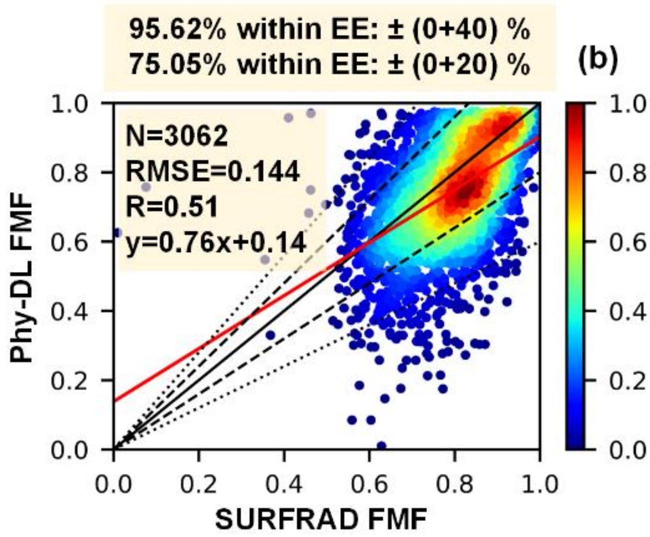

Figure R2. Phy-DL FMF at 500 nm as a function of SURFRAD FMF. The black and red solid lines are the 1:1 line and the best-fit line obtained from linear regression, respectively. The black dashed and dotted lines represent the expected error (EE) envelopes of ±20% and ±40%, respectively.

Furthermore, the Phy-DL FMF performance was validated at each SURFRAD site. **Figure R3** shows the bias of Phy-DL FMF (Phy-DL FMF minus SURFRAD FMF), retrievals falling within the ±20% EE envelope, and RMSEs at each SURFRAD site. In general, most of the sites have a mean bias and an RMSE lower than 0.1 and 0.15, respectively, with over 70% of the retrievals falling within the ±20% EE envelope. The out-of-site validation reveals that the Phy-DL FMF algorithm is reliable even in regions without AERONET sites for model training.

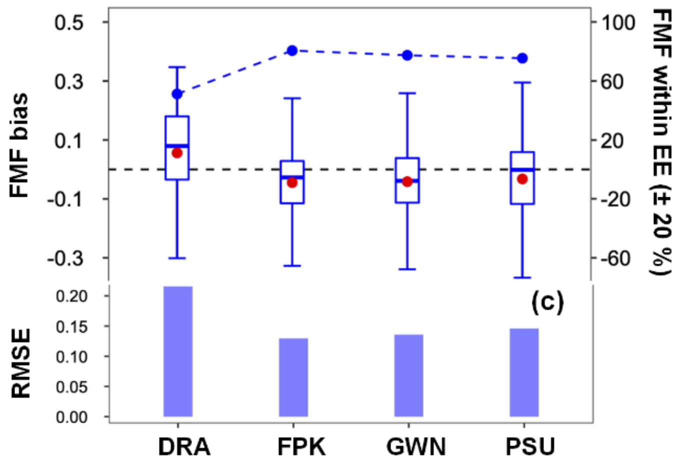

Figure R3. Boxplots of bias (Phy-DL FMF minus SURFRAD FMF), percentage of FMF estimates falling within the EE envelope of ±20% (dash-dotted line), and RMSEs at the four independent SURFRAD sites. The upper, middle, and lower lines in each box present the 75th, median, and 25th percentiles, respectively. The red point in each box represents the mean value of the FMF bias.

2. My second concern is the use of meteorological data, which are very different inputs compared to previous studies. I can see the meteorological data are widely used in fine particulate (PM2.5) retrievals because meteorology has statistical correlation and physical interaction with the PM2.5. While in this study, the author simply explained their reason as "the impact of meteorological factors", and what is this "impact" to make them use the meteorological data is not mentioned. For example, the temperature is introduced as input, I don't see the influence of temperature on FMF or how it can improve the retrieval accuracy.

Response: Thank you for this question. First, the impact of meteorological factors revealed in the significant statistical correlation between meteorological factors and FMF. We implemented the Generalized Additive Model (GAM) to fit the meteorological variables to the FMF and investigated their nonlinear relationships. **Figure R4** reveals that the meteorological variables considered, i.e., PBLH, temperature, surface pressure, RH, and wind speed, all had significant non-linear relationships with the FMF (at the 99% significance level). Both PBLH and surface pressure had similar influences on the FMF, i.e., a positive (negative) response when PBLH and surface pressure values were low (high). This is because high PBLH and surface pressure values can increase the diffusion of fine particles, decreasing the magnitude of the FMF (Tai et al., 2010). Meanwhile, the negative response of the FMF to wind speed also reflects the influence of fine-particle diffusion, as well as the contribution of dust particles strengthened by wind speed (Luo et al., 2016). Increasing temperatures corresponded to decreasing FMFs, partly due to unfavorable diffusion conditions (Tai et al., 2010). On the other hand, more fine particles are released by heating during colder seasons than during warmer seasons (Ramachandran, 2007). RH had a strong positive influence on $PM_{2.5}$ concentrations when RH was between 25% and 75%. This reflects the secondary particle formation boosted by the increasing RH, contributing to the fine particles (Tai et al., 2010).

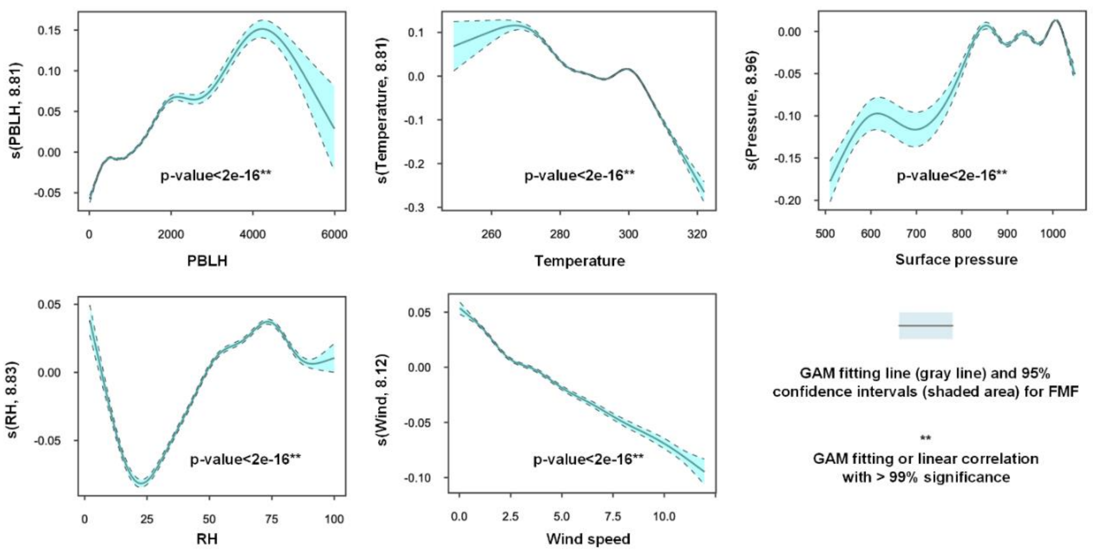

Figure R4. GAM fitting plots for the meteorological variables and the FMF. Shaded areas in the GAM plots indicate 95% confidence intervals, and the y-axes show the covariate and effective degrees of freedom of the smoothing. Asterisks (**) after each p-value indicate the 99% confidence interval of fitting.

In addition, the physical approach (SDA) does not include these potential meteorological influences. O'Neill et al. (2008) have reported that when the temperature is low, the error of the fine-mode AOD calculated by the SDA is clearly large (**Figure R5**). Although the developers of the SDA know about this issue, the relationship between meteorological factors and the FMF is complex, difficult to describe in equations. Therefore, we incorporate meteorological variables into deep learning to model these complex relationships with the FMF.

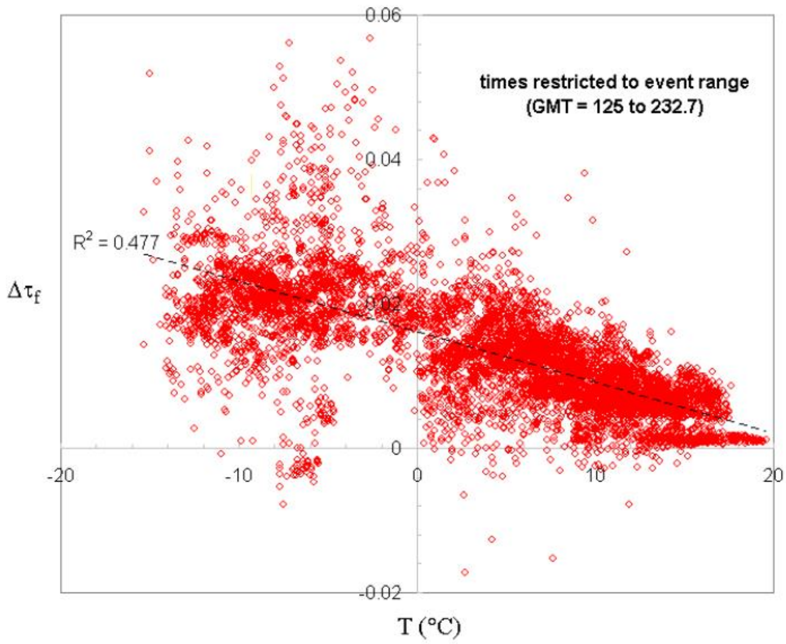

Figure R5. Variation in $\Delta\tau_f$ with detector temperature (PEARL CIMEL, May 1 to August 31, 2007). Copied from the Spectral Deconvolution Algorithm (SDA) technical memo.

Changes in the manuscript: We have revised the manuscript as follows:
(1) Added a new figure (i.e., **Figure S2)** in the supplementary file.

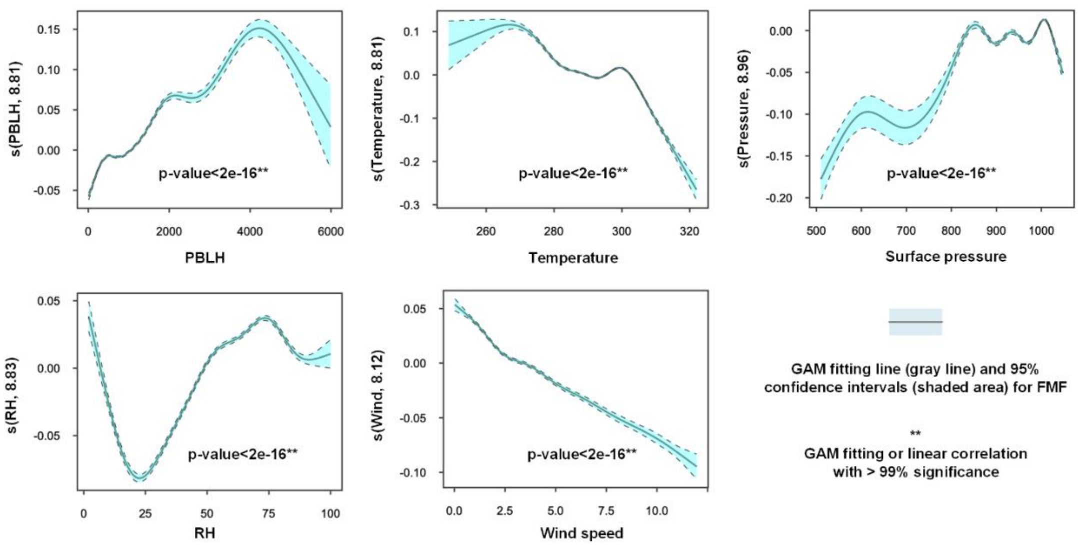

Figure S2. GAM fitting plots for the meteorological variables and the FMF. Shaded areas in the GAM plots indicate 95% confidence intervals, and the y-axes shows the covariate and effective degrees of freedom of the smoothing. The asterisks (**) after each p-value indicate the 99% confidence interval of fitting.

(2) In section 2.3 entitled "Meteorological data", we have added the following text:

Previous studies have reported that meteorological variables are significantly correlated to fine-mode and coarse-mode aerosols. Tai et al. (2010), Liang et al. (2016), and Shen et al. (2018) all revealed that meteorological variables like temperature, RH, and wind speed explain much of the variations in $PM_{2.5}$ concentrations (> 50%). Xiang et al. (2019) and Gui et al. (2019) found a negative association between BLH and $PM_{2.5}$, and Kang et al. (2014) found that fine-mode aerosols and air pressure were correlated significantly. In this study, to investigate the correlation between meteorological variables and the FMF, we implemented the Generalized Additive Model (GAM). Figure S2 reveals that the meteorological variables considered in this study, i.e., PBLH, temperature, surface pressure, RH, and wind speed, all had significant non-linear relationships with the FMF (at the 99% significance level). Both PBLH and surface pressure had similar influences on the FMF, i.e., a positive (negative) response when PBLH and surface pressure values were low (high). This is because high PBLH and surface pressure values can increase the diffusion of fine particles, decreasing the magnitude of the FMF (Tai et al., 2010). Meanwhile, the negative response of the FMF to wind speed also reflects the influence of fine-particle diffusion, as well as the contribution of dust particles strengthened by wind speed (Luo et al., 2016). Increasing temperatures corresponded to decreasing FMFs, partly due to unfavorable diffusion conditions (Tai et al., 2010). On the other hand, more fine particles are released by heating during colder seasons than during warmer seasons (Ramachandran, 2007). RH had a strong positive influence on $PM_{2.5}$ concentrations when RH was between 25% and 75%. This reflects the secondary particle formation boosted by the increasing RH that contributed to the fine particles (Tai et al., 2010). Therefore, in this study, we used surface temperature, air pressure, PBLH, RH, and wind speed as inputs to the deep-learning model.

3. For the physical model, although LUT-SDA has been used in other studies before, it is better for authors to emphasize its disadvantage with more details, rather than just listing its applications.

Response: Thank you for this valuable suggestion. Because LUT-SDA is derived based on SDA, which does not include these potential meteorological influences. While O'Neill et al. (2008) have reported that when the temperature is low, the error of the fine-mode AOD calculated by the SDA is clearly large (**Figure R5**). Although the developers of the SDA know about this issue, the relationship between meteorological factors and the FMF is complex, difficult to describe in equations.

In addition, the core of the SDA method relies on AE as input (Yan et al., 2017). The Ångström Exponent (AE) from the MODIS DT aerosol product is still highly uncertain, thus the low accuracy of AE can significantly influence the performance of the Phy-DL FMF algorithm.

Changes in the manuscript: We have revised the manuscript as follows:

In section 1 entitled "Introduction", we have added the following text:

Because LUT-SDA is derived based on SDA, which does not include these potential meteorological influences. While O'Neill et al. (2008) have reported that when the temperature is low, the error of the fine-mode AOD calculated by the SDA is clearly large. In addition, the core of the SDA method relies on AE as input (Yan et al., 2017). The Ångström Exponent (AE) from the MODIS DT aerosol product is still highly uncertain, thus the low accuracy of AE can significantly influence the performance of the Phy-DL FMF algorithm.

4. Last I think the uncertainty of this Phy-DL FMF should have a more in-depth discussion. The major content in Results is the inter-comparisons of different results in terms of different methods and FMF products, which are good, but there should be more discussion on the sources of uncertainty of this Phy-DL FMF. For example, the Figure 6 compared performance over different land types and barren land has the worst performance for all three FMFs, so the authors could discuss how the physical characteristics of barren land affected the retrieval performance.

Response: Thank you for this valuable suggestion. We discussed the sources of uncertainty of this Phy-DL FMF further.

As shown in **Figure R6,** more than 75% of the sites located on barren land have low percentages of Phy-DL FMFs (< 60%) falling within the ±20% EE envelope. About 4% of the sites have high percentages of Phy-DL FMFs (> 90%) falling within the ±20% EE envelope.

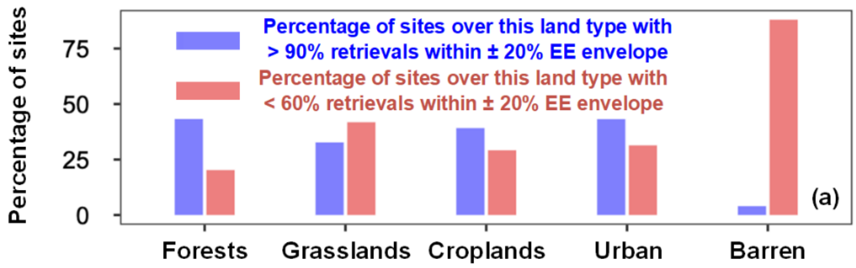

Figure R6. Bar plots of the percentage of sites with > 90% of retrievals falling within the ±20% EE envelope (blue bars) and the percentage of sites with < 60% of retrievals falling within the ±20% EE envelope (red bars) for five land types.

The barren land type is a bright surface compared to other land types where the other sites are located (**Table R1**). AODs over the bright surface used for the Phy-DL FMF retrieval were significantly overestimated, with the worst performance compared to other vegetated land-cover types (Levy et al., 2010; Petrenko and Ichoku, 2013). This suggests that the performance of the Phy-DL FMF algorithm is poor when applied to regions with barren land. **Figure R7** shows the bias of the Phy-DL FMF and the percentage of retrievals falling within the EE envelope of ±20% as a function of NDVI. As NDVI increased from < 0.1 to > 0.8, the percentage of FMF retrievals falling within the ±20% EE envelope also rose from < 70% to > 85%, and the range of bias decreased significantly. Because a higher NDVI value indicates a darker surface, **Figure R7** reveals that the Phy-DL FMF algorithm performs better over dark surfaces than over bright surfaces, resulting in a lower accuracy over the barren land type than vegetated land types.

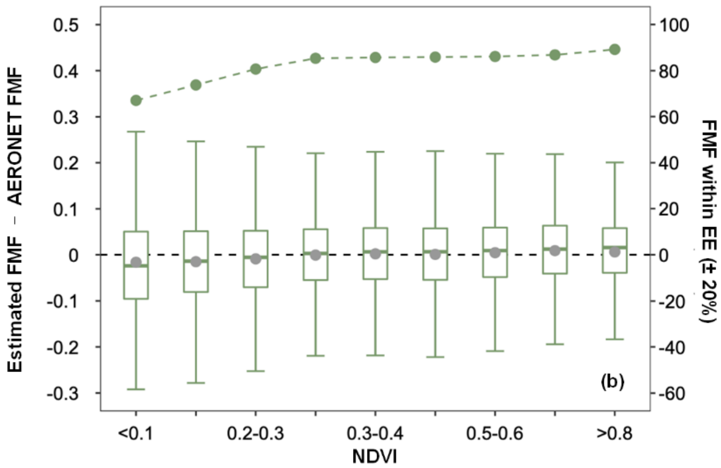

Figure R7. Box plots of the FMF bias (estimated FMF minus AERONET FMF) as a function of NDVI. The black horizontal dashed line indicates the zero bias. The gray dot in each box represents the mean value of the FMF bias. The upper, middle, and lower horizontal lines in each box show the 75th, median, and 25th percentiles, respectively. The green dots connected by the dashed curve are percentages of FMF retrievals falling within the EE envelope of ±20%.

The Ångström Exponent (AE) from the MODIS DT aerosol product is also a source of uncertainty in Phy-DL FMF. The core of the SDA method relies on AE as input (Yan et al., 2017), thus the low accuracy of AE can significantly influence the performance of the Phy-DL FMF algorithm. As shown in **Figure R8**, AEs from the MODIS MOD08 product used as input to the Phy-DL FMF algorithm performed the worst over barren land, with the highest RMSE (> 1) and the lowest percentage of retrievals falling within the EE envelope of ±0.45 (< 45%). This would result in a lower performance of the Phy-DL FMF algorithm when applied to regions with barren land.

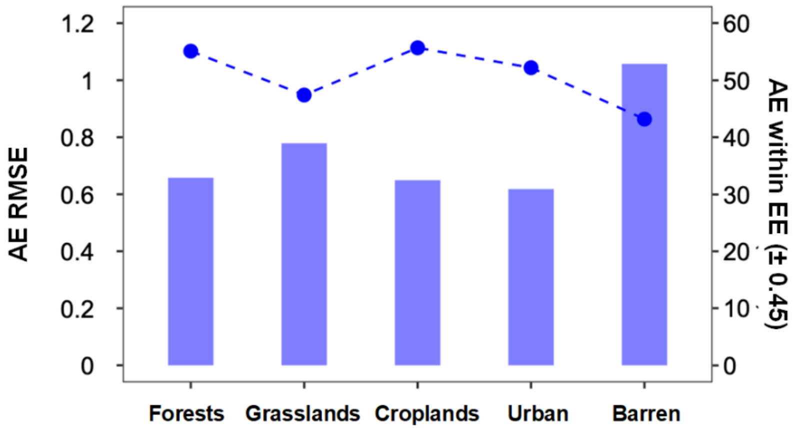

Figure R8. RMSEs (bars) and percentages of MOD08 AE falling within the EE envelope of ±0.45 (dash-dotted line) against AERONET observations for five land types. The EE envelope (±0.45) was adopted from Levy et al. (2013).

Changes in the manuscript: We have revised the manuscript as follows:

(1) Added new figures (i.e., **Figure S3** and **Figure S4)** to the supplementary file.

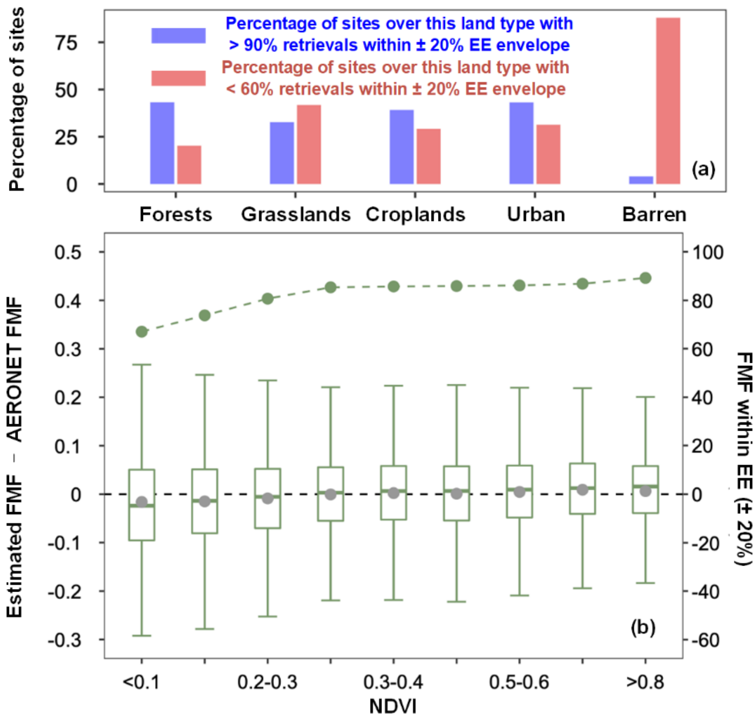

Figure S3. (a) Bar plots of the percentage of sites with > 90% of retrievals falling within the ±20% EE envelope (blue bars) and the percentage of sites with < 60% of retrievals falling within the ±20% EE envelope (red bars) for five land types. (b) Box plots of the FMF bias (estimated FMF minus AERONET FMF) as a function of NDVI. The black horizontal dashed line indicates the zero bias. The gray dot in each box represents the mean value of the FMF bias. The upper, middle, and lower horizontal lines in each box show the 75th, median, and 25th percentiles, respectively. The green dots connected by the dashed curve are percentages of FMF retrievals falling within the EE envelope of ±20%.

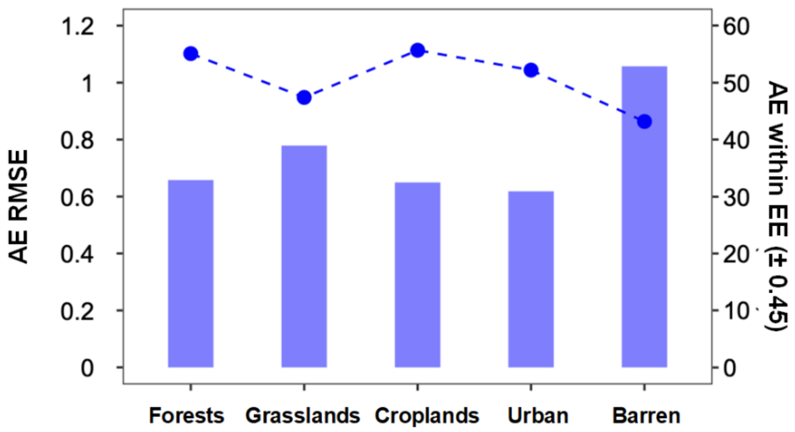

Figure S4. RMSEs (bars) and percentages of MOD08 AE falling within the EE envelope of ±0.45 (dash-dotted line) against AERONET observation for five land types. The EE envelope (±0.45) was adopted from Levy et al. (2013).

(2) In section 3.1 entitled "Phy-DL FMF validation", we have added the following text:

Figure S3a shows that more than 75% of the sites located on barren land have low percentages of Phy-DL FMFs (< 60%) falling within the EE envelope of ±20%. About 4% of the sites have high percentages of Phy-DL FMFs (> 90%) falling within the ±20% EE envelope. This suggests that the accuracy of Phy-DL FMF over barren land is much lower than over other land types. The barren land type is a bright surface compared to other land types where the other sites are located (Table S3). AODs over the bright surface used for the Phy-DL FMF retrieval were significantly overestimated, with the worst performance compared to other vegetated land-cover types (Levy et al., 2010; Petrenko and Ichoku, 2013). This suggests that the performance of the Phy-DL FMF algorithm is poor when applied to regions with barren land. Figure S3b shows the bias of the Phy-DL FMF and the percentage of retrievals falling within the EE envelope of ±20% as a function of NDVI. As NDVI increased from < 0.1 to > 0.8, the percentage of FMF retrievals falling within the ±20% EE envelope also rose from < 70% to > 85%, and the range of bias decreased significantly. The Ångström Exponent (AE) from the MODIS DT aerosol product is still highly uncertain. The core of the SDA method relies on AE as input (Yan et al., 2017). The low accuracy of AE can significantly influence the performance of the Phy-DL FMF algorithm. As shown in Figure S4, AEs from the MODIS MOD08 product used as input to the Phy-DL FMF algorithm performed the worst over barren land, with the highest RMSE (> 1) and the lowest percentage of retrievals falling within the EE envelope of ±0.45 (< 45%). This would result in a lower performance of the Phy-DL FMF algorithm when applied to regions with barren land.

Minor comments:

1. in Line 44. The "performed previously,; currently" should be "performed previously; currently"

Response: Thank you for this correction. We have corrected it in Line 44 as "performed previously; currently".

2. Figure 1. AATSR and VIIRS also provide FMF products but they were not discussed in this paper. The reason for using MODIS rather than AATSR and VIIRS should also be mentioned.

Response: Thank you for this suggestion. We added the reasons in Line 50-51 as:
In addition, Advanced Along Track Scanning Radiometer (AATSR) ended the mission in 2012 (Kolmonen et al., 2016). While VIIRS started the mission in 2012, which could provide less than 10-year global FMF product so far (Sawyer et al., 2020).

3. in Line 56. The Yonsei Aerosol Retrieval algorithm (Choi et al., 2016) is not for MODIS land-based FMF retrievals, it is for GOCI.

Response: Thank you for this correction. We deleted this irrelevant description in Line 56.

4. in Line 103. There is no direct relative humidity data ERA5. Usually it is calculated

from dew point temperature.

Response: Thank you for this correction. The relative humidity was indeed calculated from dew point temperature and air temperature, and we corrected the "relative humidity" in Line 103 as "2-m dew point temperature" and added "the relative humidity (RH) was then calculated by 2-m dew point temperature and air temperature (Tetens, 1930)."

5. in Line 104. The "ERA5" mentioned for the first time without given the full name.

Response: Thank you for this correction. We actually have explained its full name in Line 104-105 as "ERA5 is the fifth-generation product produced by the European Centre for Medium Range Weather Forecasts". And here we corrected the Line 104-105 as "obtained from the fifth-generation product produced by the European Centre for Medium Range Weather Forecasts (ERA5) (Figure S1b-f), with hourly data available since 1950 and at a 0.25° spatial resolution."

6. in Line 210. The "yr" mentioned for the first time without given the full name.

Response: Thank you for this correction. We added its full name in Line 210 as "year (yr)".

7. in 3.2. Both past and present tense showed in this part when describing the same thing. For example, "in India, FMFs are noticeably higher in autumn and winter, especially in Northern India (i.e., the Indo-Gangetic Plain), where the FMF was greater than 0.87", and they should be either past or present tense.

Response: Thank you for this correction. We thoroughly checked the tense in **3.2 Global land FMF spatial distribution and trends from 2001 to 2020** and corrected the description.

8. in Figure 6a. Why do you choose to compare the result in bins of FMF?

Response: Thank you for this question. This comparison followed the Levy et al. (2007) in **Figure R9**, which also compared different FMF products using the bins of FMF.

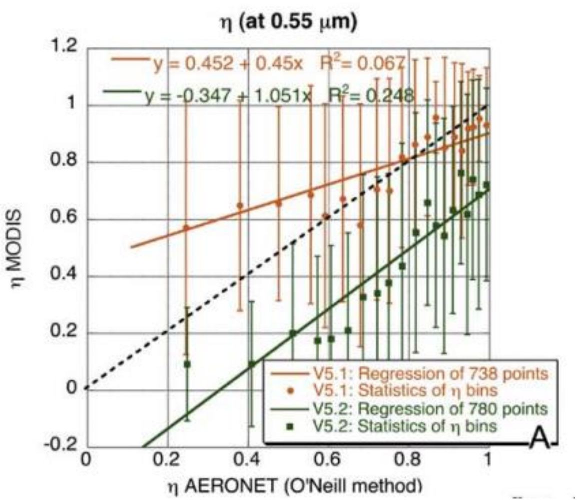

Figure R9. MODIS aerosol size retrievals compared with AERONET-derived products.

The solid shapes and error bars represent the means and standard deviations of the MODIS retrievals in 20 bins of AERONET-derived product. Retrievals from V5.1 (orange) and V5.2 (green) are shown. The regressions (solid lines) are for the clouds of all points (not shown). The η over land is retrieved at 0.55 μm and compared with AERONET η retrieved by the O'Neill et al. (2003) method. Note that η is defined differently for MODIS and AERONET and that we only show results for $\tau > 0.20$. Copied from Levy et al. (2007).

9. in Line 318 and 320. "eastern China" or "Eastern China"?

Response: Thank you for this correction. We corrected the "Eastern China" in Line 318 as "eastern China".

10. I noticed the plurals appeared randomly, for example, it is "AERONET FMF" or "AERONET FMFs". Make sure the plurals are consistent in the paper.

Response: Thank you for this correction. We thoroughly checked plurals in the manuscript and corrected the description in the manuscript.

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