

R.2 -----

(Sect 3.1)

Compared to the version of the algorithm used in Limbacher et al. (2022), the updated methodology of additionally blending the contributions of the RSA and PSA using the prescribed surface NDVI provides improved results for low-NDVI scenes. Table S4 in the supplement shows that in scenes where prescribed-surface NDVI < 0.1 the new CSA displays an RMSE 45% lower than the previous version, with a 43% lower bias. In the regime of 0.1 < NDVI < 0.25, the new CSA also shows improved RMSE (0.089 vs 0.114), and bias (0.001 vs 0.014).

**Table S4:** Results of the new MISR CSA using additional attenuation depending on NDVI introduced in this paper alongside the version used in Limbacher et. al., 2022, stratified according to three bins of scene-wide prescribed-surface NDVI. N is the number of coincidences, RMSE is the root mean squared error, MAE is median average error, bias is the mean AERONET-MISR bias, and r is the Pearson correlation coefficient.

	New CSA	CSA (2022)
	NDVI < 0.10	
N	6942	6942
RMSE	0.125	0.229
MAE	0.08	0.103
bias	0.084	0.147
r	0.917	0.889
	0.10 < NDVI < 0.25	
N	11513	11513
RMSE	0.089	0.114
MAE	0.032	0.033
bias	0.001	0.014
r	0.948	0.917
	NDVI > 0.25	
N	57344	57344
RMSE	0.082	0.083
MAE	0.026	0.026
bias	0	0.001
r	0.946	0.943

R.3 -----

(Sect. 3.1)

Table S5 in the supplement constrains MISR vs. AERONET AOD to scenes where PSA AOD > 1.0. These results display the low bias of the RSA at high AOD, and that the CSA achieves lower absolute bias (-0.029 vs. -0.152) than the RSA by leveraging the results of the PSA.

**Table S5:** Statistics for MISR over-land RSA, PSA and CSA 550nm AOD vs AERONET 550nm AOD, filtered by MISR scene-wide PSA AOD > 1. N is the number of coincidences, RMSE is root mean squared error, MAE is mean average error, bias is the mean MISR – AERONET bias, and r is the Pearson correlation coefficient.

	RSA	PSA	CSA
N	1861	1861	1861
RMSE	0.387	0.508	0.299
MAE	0.168	0.286	0.143
bias	-0.152	0.217	-0.029
r	0.847	0.683	0.880

R.5 -----

(Sect 3.4)

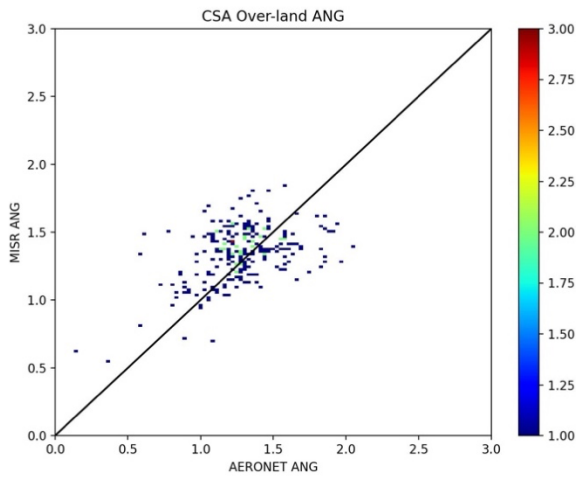
A limited number of sites in meteorological winter were selected to assess the performance of the MISR CSA in scenes likely at times to contain significant urban and/or industrial pollution. Table S2.3 in the supplement contains the list of sites used in this analysis. Figure S7 and Table S11 display results for MISR CSA vs AERONET Ångström exponent (446-867nm) for this selection of sites, constrained to MISR AOD > 0.2. With 240 coincidences, Fig. S7 shows incident data primarily grouped around  $1.0 < \text{ANG} < 2.0$  for both MISR and AERONET, which would be expected for sites primarily dominated by small, urban and/or industrial aerosols. For these coincidences, MAE is 0.166 and RMSE is 0.260, with a bias of 0.062.

Figure S8 and Table S12 display results for MISR vs AERONET FMF and SSA for the selection of sites in Table S2.3 and constrained for MISR AOD > 0.2, with cases for MISR AOD  $\geq 0.5$  highlighted in a different color. The number of AERONET almucantar coincidences for this selection is 203. For FMF, RMSE is 0.096, MAE is 0.058, with a bias of -0.030. For the SSA analysis, RMSE is 0.040, MAE is 0.021 with a bias of 0.013 (Table S12). Most coincidences for FMF in Fig. S8 are grouped towards being fine-mode dominated, and the SSA for these data is primarily between 0.85 and 0.95, as might be expected for pollution particles.

**Table S2.3:** AERONET sites and months selected for likely industrially-polluting sources

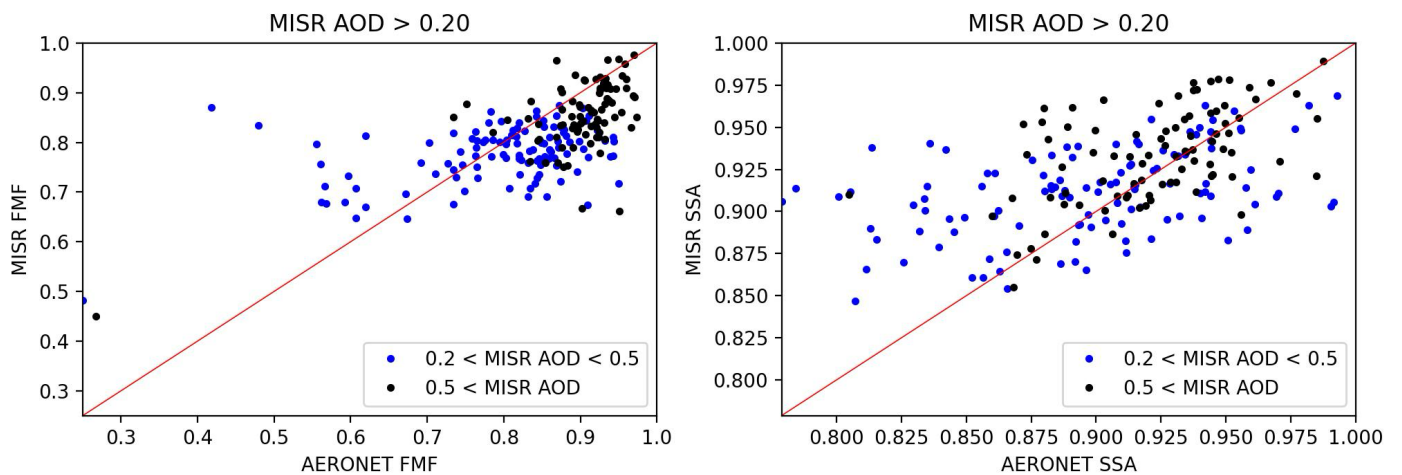
Janpur	D12
Beijing	D12
London	D12
Mexico City	D12

**Figure S7:** Plot of over-land MISR CSA vs AERONET Ångström exponent (446-867nm) for sites selected to be likely dominated by urban and/or industrial pollution aerosol sources.



**Figure S8:** Plots of over-land MISR CSA vs AERONET fine-mode AOD<sub>550</sub> fraction (FMF) and 550nm single-scattering-albedo (SSA) for over-land sites likely to contain urban and/or industrial pollution aerosol sources. Results are color-coded, with blue points corresponding to cases where  $0.2 < \text{CSA AOD} < 0.5$ , and black points corresponding to cases for which  $\text{CSA AOD} \geq 0.5$ .

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**Table S11:** Statistics for MISR CSA vs Ångström exponent (446-867nm) for sites selected to be likely dominated by urban and/or industrial pollution aerosol sources, constrained to MISR CSA AOD > 0.2. Statistics correspond to data in Fig. S7.

N	240
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r	0.448
RMSE	0.26
MAE	0.166
bias	0.062