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Supplement of

Measurement-based climatology of aerosol direct radiative effect, its sensitivities, and uncertainties from a background southeast US site

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Supplemental Materials

Overview

The following materials supplement the manuscript “*Measurement-based climatology of aerosol direct radiative effect, its sensitivities, and uncertainties from a background southeastern US site*”. We first apply techniques identical to those described in Sects. 4.3 and 4.4 for calculating sensitivities and measurement uncertainties in the aerosol direct radiative effect (DRE) to calculate the sensitivities and uncertainties in aerosol radiative efficiency (RE; equal to the DRE per unit aerosol optical depth) at 550 nm (Sects. S1 and S2). We then evaluate the sensitivity of DRE to surface types (Sect. S3), which facilitates comparisons with other studies in Sect. 5.2 of the paper.

S1. Sensitivity of RE to aerosol optical properties and surface reflectance

The uncertainty ΔRE follows an equation identical to Eq. (7) for ΔDRE

$$\begin{aligned} \Delta RE^2 = & s_{AOD}^2 \Delta AOD^2 + s_{\omega_0}^2 \Delta \omega_0^2 + s_g^2 \Delta g^2 + s_R^2 \Delta R^2 + 2s_{AOD}s_{\omega_0} \text{corr}(AOD, \omega_0) \Delta AOD \cdot \Delta \omega_0 \\ & + 2s_{AOD}s_g \text{corr}(AOD, g) \Delta AOD \cdot \Delta g + 2s_{AOD}s_R \text{corr}(AOD, R) \Delta AOD \cdot \Delta R + 2s_{\omega_0}s_g \text{corr}(\omega_0, g) \Delta \omega_0 \cdot \Delta g + \\ & 2s_{\omega_0}s_R \text{corr}(\omega_0, R) \Delta \omega_0 \cdot \Delta R + 2s_g s_R \text{corr}(g, R) \Delta g \cdot \Delta R \end{aligned} \quad (S1)$$

We use lower-case ‘s’ for the RE sensitivities, so as not to confuse them with the upper-case ‘S’ used in the main paper for the DRE sensitivities. We use the same base case aerosol optical properties and surface reflectance (Table 1), measurement uncertainties (Table 2), and correlations amongst aerosol optical properties (Table 3) for calculating the RE sensitivities and uncertainties as for DRE sensitivities (Sect. 5.2) and uncertainties (Sect. 5.3).

Aerosol RE is most sensitive to single-scattering albedo (ω_0) for all seasons (Table S1) and is least sensitive to aerosol optical depth (AOD). For the most part, sensitivities of RE to scattering asymmetry parameter (g) are comparable and surface reflectance (R). For all seasons except summer, RE is ~1.5 times more sensitive to ω_0 than to g at the top-of-atmosphere (TOA) and ~2.5 times more sensitive to ω_0 than to g at the surface. In summer, RE sensitivity to ω_0 is higher than sensitivity to g by a factor of ~2 at the TOA and ~3 at the surface. The seasonal variations in s_{ω_0} , s_g , and s_R (Table S1) are much smaller than those of S_{ω_0} , S_g , and S_R (Tables 5 and 6), with all varying by a factor of 2 or less with season. The fact that s_{ω_0} , s_g , and s_R all have the same sign (positive) at the surface (Table S1) is what drives surface RE to such small negative values in the

summer (Fig.3d of main paper), when g , ω_0 , and R are all highest and to more negative values in winter, when g , ω_0 , and R are all lowest. The larger sensitivity of RE to AOD during DEC and MAR is consistent with the slight departure from linearity of the DRE versus AOD curve during these months (Figs. 5a and 6a). By definition, RE is the DRE per unit AOD so the sensitivity s_{aod} is the second derivative of DRE with respect to AOD.

S2. Uncertainties in RE

As with the uncertainties in DRE (Sect. 5.3), the RE uncertainties at the TOA (Table S2) and surface (Table S3) are largely influenced by the sensitivities (Table S1). The uncertainties in ω_0 are the largest contributor to ΔRE during all seasons, especially at the surface. Inclusion of covariances amongst aerosol optical properties increases ΔRE by $\sim 20\text{-}25\%$ at the TOA and $\sim 10\text{-}15\%$ at the surface, for all seasons. Still, the RE uncertainties are relatively small for all seasons, ranging from 11-17% of RE computed using base-case properties (Table 1) at the TOA.

S3. Dependence of DRE on AOD and ω_0 for different surface types

Aerosol DRE depends not only on the aerosol optical properties (AOD, ω_0 , g) but also on the underlying surface, namely its spectral reflectivity. In the main paper, we parameterized the wavelength-dependent surface reflectance by its spectrally-averaged value R . We also parameterized it as a linear combination of the spectral reflectance curves produced by the surface types (snow, water, sand, vegetation) used as inputs to the SBDART radiative transfer model. In this section, we plot the dependence of DRE on AOD and ω_0 (the two most influential aerosol parameters; Sect. 5.2) for each of the individual surface types. The results will facilitate comparisons with other studies (Gadhavi and Jayaraman, 2004) and help illustrate how the relative albedos of the aerosol and underlying surface impact DRE.

Table S1. Sensitivity of aerosol radiative efficiency (RE) at the top-of-atmosphere (TOA) and surface to AOD, ω_0 , g, and R, which are used to calculate the RE uncertainties. Sensitivities $S_i = \frac{\partial RE}{\partial \rho_i}$ are calculated as the slope of RE versus ρ_i , evaluated at base case values (Table 1). All sensitivities are in units of $W m^{-2} AOD^{-1}$ per unit change in the parameter ρ_i . The correlations between aerosol optical properties are used along with uncertainties (Table 2) to calculate the covariances used in the RE uncertainty calculations (Tables S2 and S3).

Property ρ_i	MAR	JUN	SEP	DEC
TOA s_{AOD}	35.1	19.8	20.9	47.0
TOA s_{ω_0}	-182	-258	-178	-132
TOA s_g	118	121	116	91.8
TOA s_R	158	113	91	102
Surface s_{AOD}	34.8	18.9	20.8	55.6
Surface s_{ω_0}	310	357	301	214
Surface s_g	122	126	120	92.8
Surface s_R	130	93.2	76.9	89.3
Corr(AOD, ω_0)	-0.02	0.25	0.57	0.10
Corr(AOD, g)	-0.08	0.30	0.56	0.15
Corr(ω_0 , g)	0.78	0.79	0.85	0.84

Table S2. Calculated uncertainties in RE at the TOA, using the sensitivities and correlations given in Table S1 and measurement uncertainties given in Table 2 as inputs to Eq.S1. Units of ΔRE are $W m^{-2} AOD^{-1}$.

	MAR	JUN	SEP	DEC
ΔRE_{AOD}	0.35	0.20	0.21	0.47
ΔRE_{ω_0}	5.5	5.2	3.6	3.9
ΔRE_g	1.2	1.2	1.2	0.92
ΔRE_R	3.2	2.3	1.8	2.0
Sum of covariance terms	9.9	10	8.1	6.6
ΔRE	5.6	5.3	3.8	4.1
(covariance terms not included)				
ΔRE	7.2	6.6	5.0	5.2
(covariance terms included)				
RE (Base case)	-48.6	-38.1	-36.4	-45.8
$\Delta RE^a / RE$ (Base Case)	0.15	0.17	0.14	0.12

^aUncertainty includes covariance terms

Table S3. Calculated uncertainties in RE at the surface, using the sensitivities and correlations given in Table S1 and measurement uncertainties given in Table 2 as inputs to Eq.S1. Units of Δ DRE are $W m^{-2}AOD^{-1}$

	MAR	JUN	SEP	DEC
ΔRE_{AOD}	0.35	0.19	0.21	0.56
$\Delta RE_{\omega 0}$	9.3	7.1	6.0	6.4
ΔRE_g	1.2	1.3	1.2	0.93
ΔRE_R	2.6	1.9	1.5	1.8
Sum of covariance terms	17	15	14	11
ΔRE (covariance terms not included)	9.7	7.5	6.3	6.7
ΔRE (covariance terms included)	11	8.4	7.4	7.5
RE (Base case)	-91.8	-72.0	-62.9	-75.3
$\Delta RE^a / RE$ (Base Case)	0.12	0.17	0.17	0.11

^aUncertainty includes covariance terms

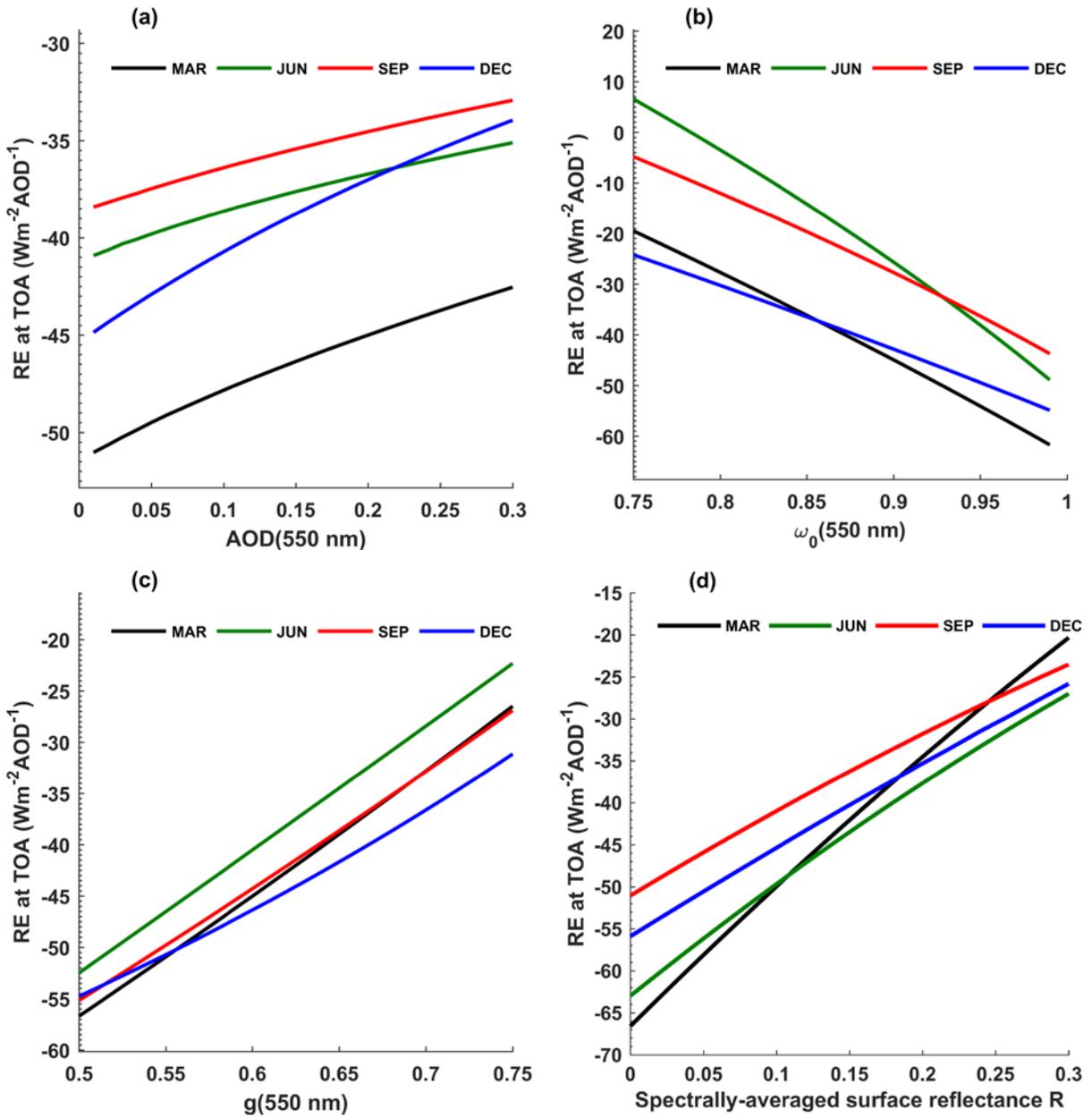


Figure S1. Seasonal dependence of the sensitivity of top-of-atmosphere (TOA) aerosol RE to (a) AOD; (b) ω_0 ; (c) g; and (d) spectrally-averaged surface reflectance R

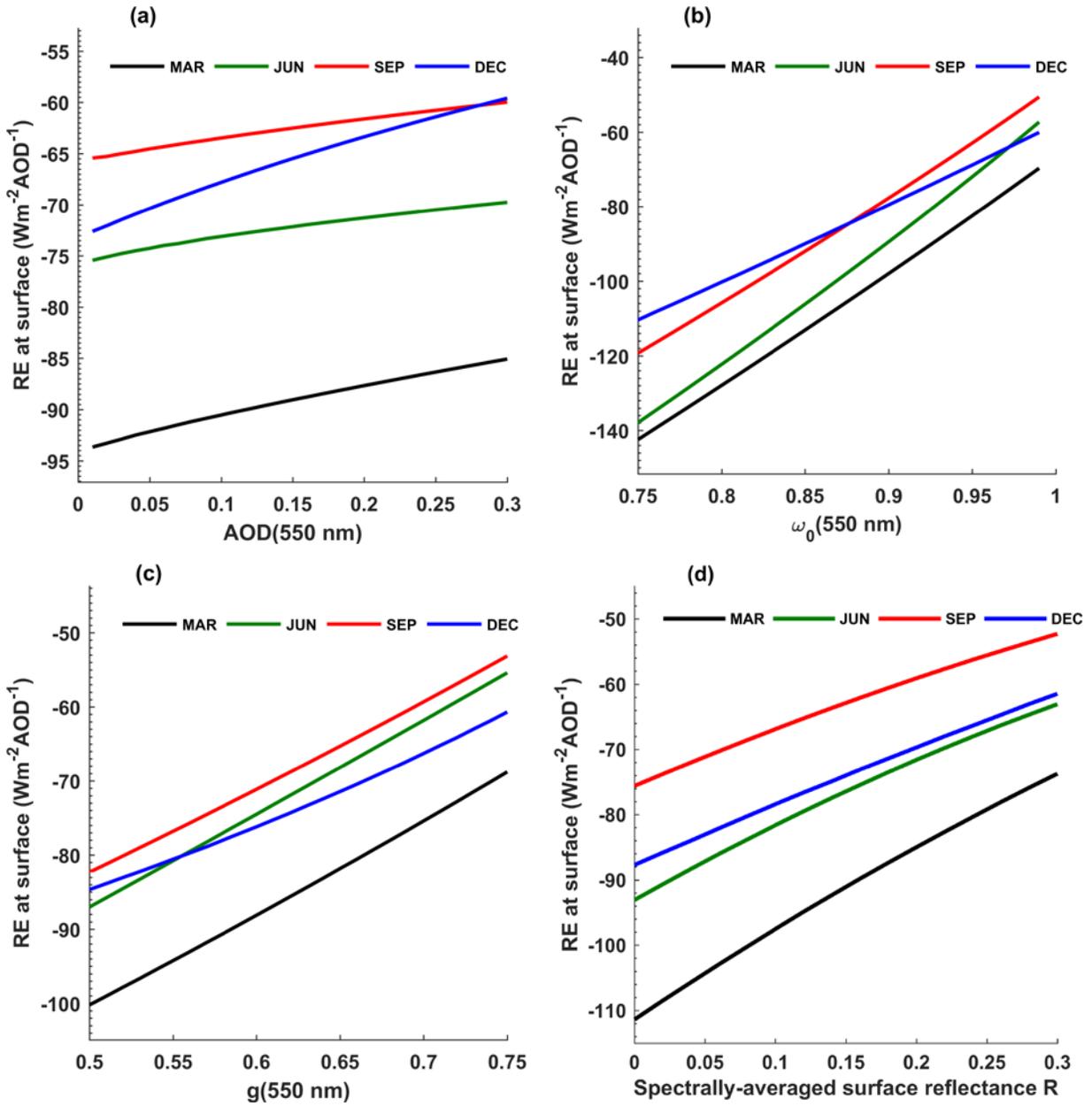


Figure S2. Seasonal dependence of the sensitivity of aerosol RE at the surface to (a) AOD; (b) ω_0 ; (c) g ; and (d) spectrally-averaged surface reflectance R

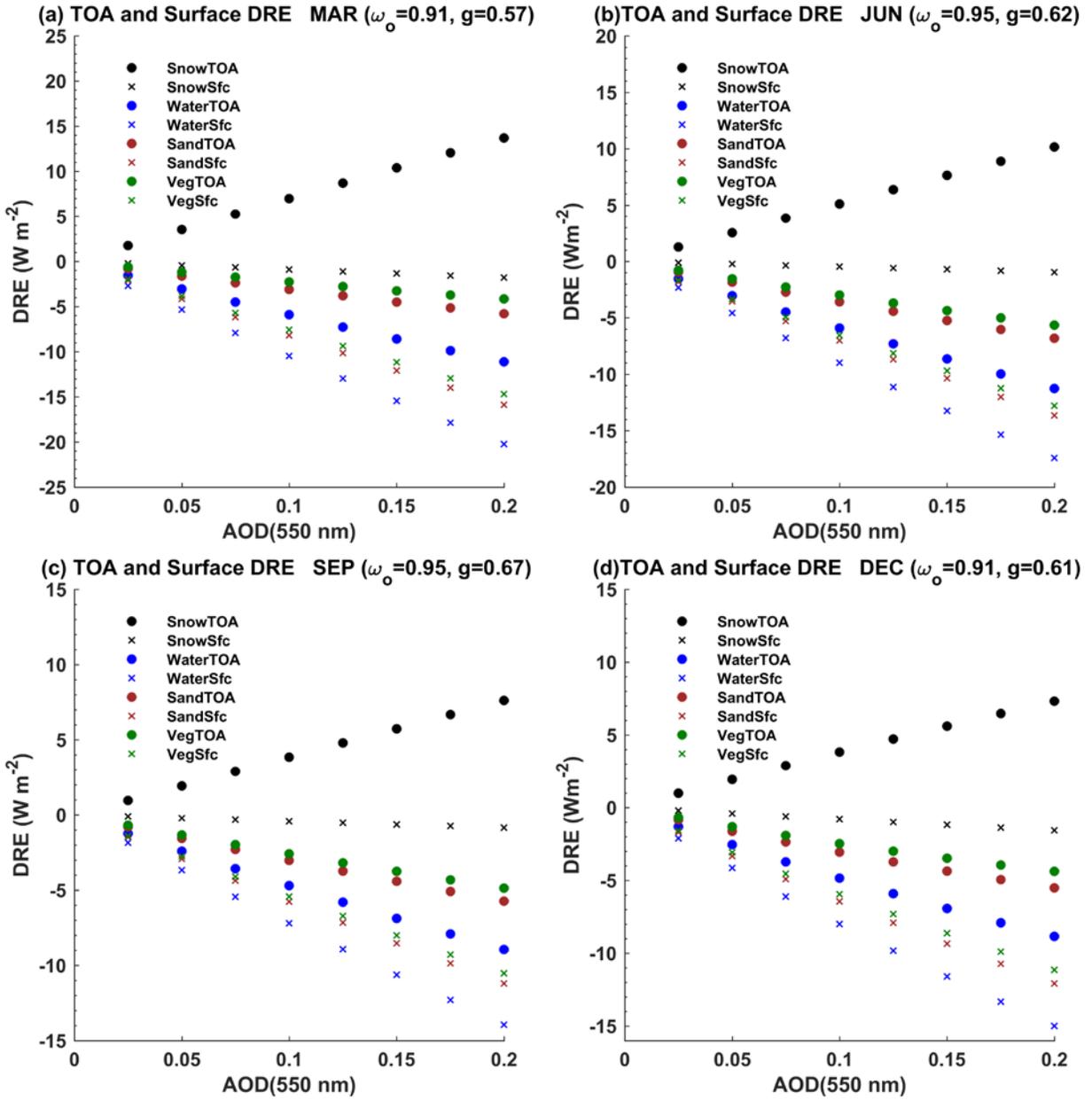


Fig. S3. Sensitivity of DRE at the TOA and surface to changes in AOD for the following surface types: snow, seawater, sand, and vegetation; for MAR, JUN, SEP, and DEC. The base-case value of ω_0 and g for each season (Table 1 of paper) are included in each plot title.

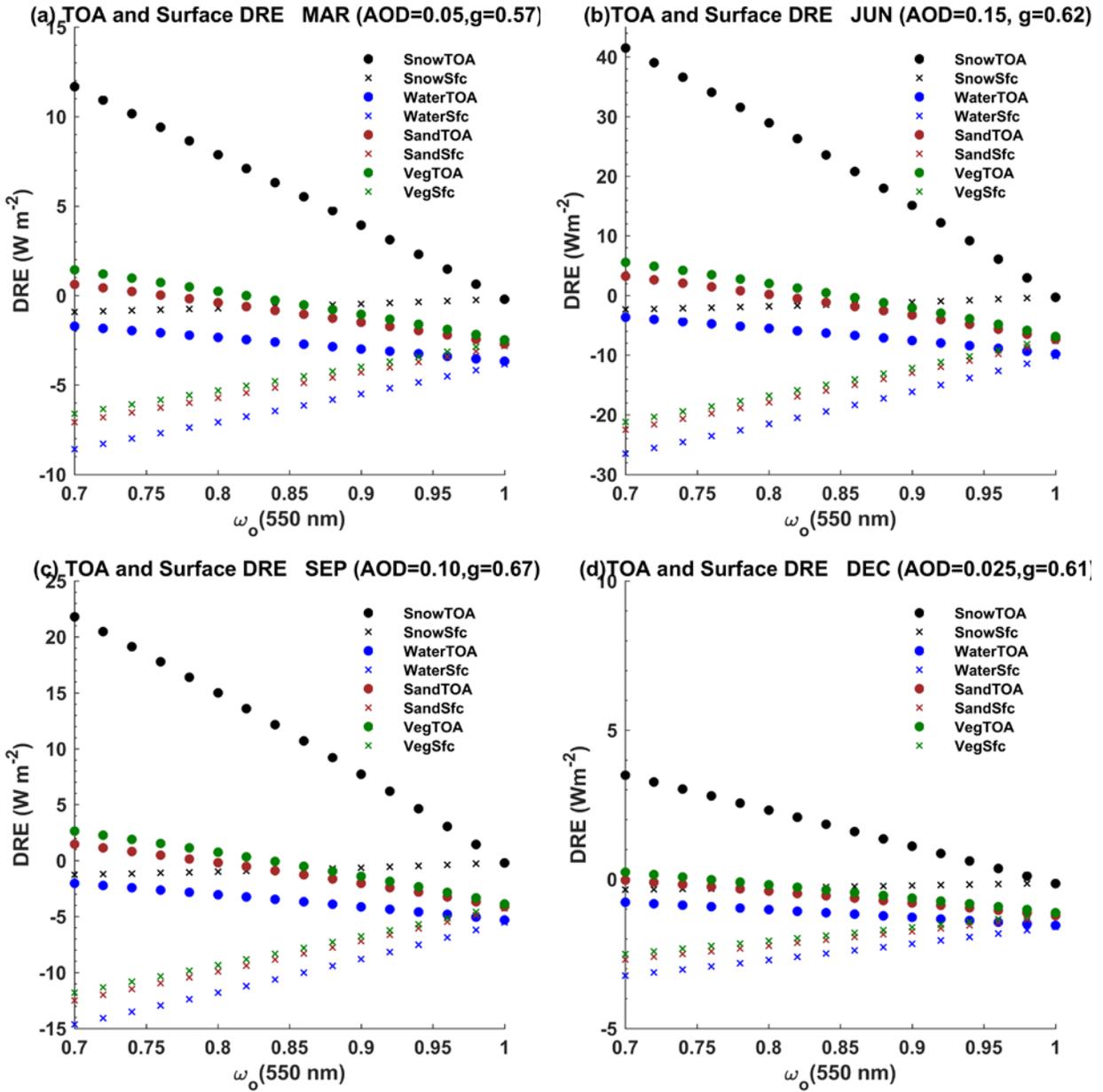


Fig. S4. Sensitivity of DRE at the TOA and surface to changes in ω_0 for the following surface types: snow, seawater, sand, and vegetation; for MAR, JUN, SEP, and DEC. The base-case value of AOD and g for each season (Table 1 of paper) are included in each plot title.