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”. Techniques identical to those described in Sects. 4.3 and 4.4 for calculating sensitivities and uncertainties in the aerosol direct radiative effect (DRE) can be used to calculate the same for aerosol radiative efficiency (RE). The uncertainty ΔRE follows an equation identical to Eq. (7) for ΔDRE

$$\Delta RE^2 = s_{AOD}^2 \Delta AOD^2 + s_{\omega_0}^2 \Delta \omega_0{}^2 + s_g^2 \Delta g^2 + s_R^2 \Delta R^2 + 2 s_{AOD} s_{\omega_0} corr(AOD, \omega_0) \Delta AOD \cdot \Delta \omega_0$$
$$+ 2 s_{AOD} s_g corr(AOD, g) \Delta AOD \cdot \Delta g + 2 s_{AOD} s_R corr(AOD, R) \Delta AOD \cdot \Delta R + 2 s_{\omega_0} s_g corr(\omega_0, g) \Delta \omega_0 \cdot \Delta g +$$
$$2 s_{\omega_0} s_R corr(\omega_0, R) \Delta \omega_0 \cdot \Delta R + 2 s_g s_R corr(g, R) \Delta g \cdot \Delta R$$
(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).

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

Aerosol RE is most sensitive to single-scattering albedo (ω_0) for all seasons (Table S1), followed by scattering asymmetry parameter (g). It is least sensitive to aerosol optical depth (AOD) 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 $s_{\omega 0}$, s_g, and s_R (Table S1) are much smaller than those of $S_{\omega 0}$, S_g, and S_R (Tables 5 and 6), with all varying by a factor of 2 or less with season. The fact that $s_{\omega 0}$, 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 proportional to 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 ~20-25% at both the TOA and surface, for all seasons. Still, the RE uncertainties are relatively small for all seasons, ranging from 6-13% of RE computed using base-case properties (Table 1) at the TOA and 5-9% at the surface. While fractional uncertainty in DRE is highest in winter (Tables 5 and 6), that of RE is lowest in winter (Tables S2 and S3).

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 p_i	MAR	JUN	SEP	DEC
TOA s_{AOD}	35.1	19.8	20.9	47.0
TOA $s_{\omega 0}$	-182	-258	-178	-132
TOA s_g	118	121	116	91.8
TOA s_R	18.3	22.7	13.8	11.0
Surface s_{AOD}	34.8	18.9	20.8	55.6
Surface $s_{\omega 0}$	310	357	301	214
Surface s_g	122	126	120	92.8
Surface s_R	15.3	18.3	11.4	10.0
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
$\Delta RE_{\omega 0}$	2.7	3.9	2.7	2.0
ΔRE_{g}	1.2	1.2	1.2	0.92
ΔRE_{R}	0.37	0.44	0.28	0.22
Sum of covariance terms	4.9	7.9	6.2	3.4
ΔRE (covariance terms not included)	3.0	4.1	2.9	2.2
ΔRE (covariance terms included)	3.7	5.0	3.8	2.9
RE (Base case)	-48.6	-38.1	-36.4	-45.8
ΔRE[a] / RE (Base Case)	0.077	0.13	0.11	0.063

[a]Uncertainty 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 ΔDRE are W $m^{-2}AOD^{-1}$

	MAR	JUN	SEP	DEC
ΔRE_{AOD}	0.35	0.19	0.21	0.56
$\Delta RE_{\omega 0}$	4.6	5.4	4.5	3.2
ΔRE_{g}	1.2	1.3	1.2	0.93
ΔRE_{R}	0.31	0.37	0.23	0.20
Sum of covariance terms	8.7	11	11	5.5
ΔRE (covariance terms not included)	4.8	5.5	4.7	3.4
ΔRE (covariance terms included)	5.7	6.5	5.7	4.1
RE (Base case)	-91.8	-72.0	-62.9	-75.3
ΔRE[a] / RE (Base Case)	0.062	0.090	0.090	0.055

[a]Uncertainty includes covariance terms