

**Reply to Reviewer #1-comments for Atmos. Chem.Phys. Discuss.,
<https://doi.org/10.5194/acp-2017-513>, Measurement-based climatology of aerosol direct radiative effect, its sensitivities, and uncertainties from a background southeast U.S. site, by JP Sherman and A. McComiskey**

We thank anonymous reviewer #1 for her/his excellent suggestions, particularly those related to the Analysis/Discussion section. We've gone to great lengths to implement nearly all of the suggestions made by both reviewers and believe that these changes have significantly improved the paper. We structure our responses to each reviewer comment/suggestion as follows: (1) Reviewer 2 Comment xx, where xx is the comment number; (2) Authors' response; and (3) Changes to Paper.

Reviewer 1 Comment 1: As a suggestion for improvement. The manuscript focuses a great deal on sensitivities to AOD, single scattering albedo (SSA), asymmetry parameter (g) and surface reflectance (R). I believe sensitivities are intrinsic property of radiative transfer model and their emphasis would have made more sense if the results were about comparison of different radiative transfer models. In the present case, their findings on sensitivities will differ from others (previous studies) only to extent differences in base cases and impact of non-linearity over the range of difference. I believe emphasis should have been more on seasonal variations in aerosol properties and how they differ from generic aerosol models used in various models and satellite retrieval algorithms, and ultimately what would be the penalty in terms of error in DRE if the generic models are used instead of measurements.

Authors' Response: We respectfully disagree with the reviewer's assertion that the sensitivities are intrinsic property of the RTM (if we are correctly interpreting her/him) and point to the results from two studies. As part of a radiative transfer closure study, Michalsky et al.(2006) found that six radiative transfer models (RTMs) were all able to simulate clear-sky direct and diffuse shortwave fluxes to within 1.0% and 1.9%, respectively, of the measured fluxes, provided that all models used the same aerosol inputs. They concluded that the largest source of difference in the RTM outputs is likely due to how the RTM extrapolates the aerosol optical properties used as inputs (particularly AOD) to unspecified wavelengths. As a follow-up to this study, McComiskey et al. (2008) showed that the sensitivities of clear-sky DRE to changes in aerosol inputs were not dependent on the model used. Both studies demonstrate that the RTMs are capable of calculating clear-sky DRE with high precision and that DRE uncertainty arises largely from incorrectly-specified aerosol optical properties

The reviewer is likely correct in her/his assessment that the sensitivities are primarily dependent on base-case aerosol optical properties (and not on the model used) and this in fact is why regionally-representative aerosol measurements possessing low uncertainties (such as from NOAA-ESRL and AERONET sites) are needed to improve DRE estimates. Fortunately, studies (Sherman et al., 2015; Delene and Ogren 2002; and others) have shown that intensive aerosol optical properties such as SSA are not too different between many North American regions and AOD has decreased significantly over much of North America in the past 2 decades. As a result, the results in this paper should be applicable over at least the SE US and likely much of eastern continental North America. The technique can also be easily extended to industrial regions

where the sensitivity values may not be applicable, given a co-located NOAA-ESRL/AERONET site (ex: Bondville, IL; Egbert, Ontario; etc).

Changes to Paper: We have added the following text to the first paragraph of the Introduction section: *“As part of a recent radiative transfer closure study, Michalsky et al.(2006) found that six radiative transfer models (RTMs) were all able to simulate the observed clear-sky direct and diffuse shortwave fluxes to within 1.0% and 1.9%, respectively, of the measured fluxes, provided that all models used the same aerosol inputs. They concluded that the largest source of difference in the RTM-derived fluxes is likely due to how the RTM extrapolates the aerosol optical properties used as inputs (particularly AOD) to unspecified wavelengths. As a follow-up to this study, McComiskey et al. (2008) showed that the sensitivities of clear-sky DRE to changes in aerosol inputs was not dependent on the model used. Both studies demonstrate that the RTMs are capable of calculating clear-sky DRE with high precision and that DRE uncertainty arises largely from incorrectly-specified aerosol optical properties, which can result from lack of regionally-representative aerosol measurements, measurement uncertainties, and spatio-temporal aerosol variability.”*

Reviewer 1 Technical Comment 1: Authors discuss effect of measurement uncertainties on uncertainties in DRE. However, it is rare that DRE is estimated for instantaneous values measured by various instruments. Generally, required parameters are averaged over certain time-period (typically one day) and will have associated variabilities, quite often larger than instrumental error leading to further uncertainty in DRE estimation. I am not clear about whether authors mention uncertainty in DRE including variability of input parameters or only of measurement error. Authors may consider including discussion on uncertainty that arises from variability of the input parameters in addition to the measurement error.

Authors' Response: The reviewer brings up a very good point, namely that diurnal variability in the aerosol optical properties serving as inputs to the radiative transfer model (AOD, SSA, g) can often lead to DRE uncertainties that are at least as large as DRE uncertainties due to measurement uncertainties. While the main purpose of our paper is to quantify uncertainties in DRE due to the aerosol measurement uncertainties (which likely leads to a lower bound in DRE uncertainty), we do agree that some discussion of diurnal aerosol variability and its effect on DRE calculations is warranted.

Changes to Paper: We have added a short section (Sect. 5.4) to the manuscript, discussing DRE uncertainties due to diurnal aerosol variability. We apply the DRE sensitivity parameters (Sect. 5.2) along with an estimate of aerosol diurnal variability, to estimate diurnally-averaged DRE uncertainties due to diurnal aerosol variability. To estimate diurnal aerosol variability, we apply the method used by Sherman et al (2015) and Sherman et al. (2016), both of which are referenced in the manuscript. For each season, we form hourly averages of all AOD, SSA, and g values at 550nm. We then bin the values by hour of day and form statistics for each hour of the day (mean, standard error of the mean). We also form statistics using all hours of the day (i.e. the entire dataset for that season). We include a new figure (Figs. 8(a)-(c)), containing plots of the diurnal cycle of mean AOD, SSA, and g at 550nm for each season. We include error bars for each hour to indicate confidence in the mean values (i.e. standard error of the mean) and to assess whether the diurnal variability in mean aerosol properties is statistically-significant. We

estimate “diurnal variability” of each aerosol input (AOD, SSA, and g), using the difference between the diurnally-averaged values and the mean values for individual hours of the day. For example, suppose that the daily-mean SSA at 550nm during summer is 0.96 and that the mean SSA values for individual hours of the day ranged from 0.94 to 0.98, we would estimate the peak error in using the daily-averaged SSA (0.96) as $\Delta\text{SSA}=0.02$. Use of the peak error leads to upper bounds on the resulting DRE uncertainty estimates but represent a simple application of the DRE sensitivity parameters to estimate DRE uncertainties. We report the DRE uncertainties due to diurnal aerosol variability in a newly-created table (Table 6). We also preface the phrase “DRE uncertainties” with the word “measurement” throughout the paper, in cases where confusion may exist.

Reviewer 1 Technical Comment 2: Authors have used power law equation to extrapolate AOD and SSA beyond visible wavelength. Originally, the power law was derived for visible wavelength range and there aren’t many evidences to suggest applicability of the law in infrared. At the same time, I believe authors may not have made big error in DRE numbers in doing so as the solar energy in that part of spectrum is very little compared to visible range. However, I feel a caveat in the manuscript is necessary to reflect that power law assumption may or may not be valid in infra-red region of the spectrum.

Authors’ Response: We agree completely with the reviewer and this comment is supported by the Michalsky et al., 2006 study (See response to Comment 1 above). We have added a caveat to this extent.

Changes to Paper: We have added the following passage to the first paragraph of Sect. 3.1- Aerosol Optical Properties: *“We note that the power-law expressions (Eqs. 1,2, and 4) used to extrapolate aerosol properties measured largely at visible wavelengths to the infra-red may or may not represent their true spectral dependence. However, the solar flux in the infra-red is much less than that in the visible so simple aerosol spectral parameterizations should be sufficient for broadband DRE calculations.”*

Reviewer 1 Technical Comment 3: Authors imply on page 10(line 2 to 4) that uncertainty in SSA at higher RH is not known. However in the section 3, authors have mentioned that the site is equipped with scanning humidograph to study effect of RH on scattering and absorption coefficient. Authors may explain why can’t this data be used to find uncertainty in SSA at high RH?

Authors’ Response: A scanning humidograph (Sheridan, et al., 2001) is employed at APP to measure the RH dependence of scattering and hemispheric backscatter coefficients (σ_{sp} and σ_{bsp}) but not absorption coefficient. Radiative transfer models typically only treat the scattering dependence of RH, and assume that absorption changes negligibly with RH. While this approach may or may not hold true for all aerosol types (ex: some organics, sulfur-coated soot), the dependence of absorption on RH is experimentally very difficult for all but laboratory studies (especially at high RH) under very controlled conditions (Brem et al., 2012) and is ignored in our calculations. Thus, we only correct the scattering coefficient to ambient RH in our corrections of

SSA. Estimates of the uncertainties in hygroscopic dependence of light scattering coefficient σ_{sp} are scarce and depend primarily on the uncertainties in RH and in nephelometer-measured scattering coefficient, in addition to system-dependent particle losses in the humidograph. One study (Titos et al. (2016)) estimates the uncertainty in hygroscopic σ_{sp} enhancement for humidographs similar to that deployed at APP and we now propagate this uncertainty through the calculations to estimate uncertainty in SSA.

Changes to Paper:

1. We clarify how the humidograph corrects scattering and backscattering coefficients to ambient RH by adding the following text to Sect. 3.1-Single Scattering Albedo and Scattering Asymmetry Parameter: *“The humidograph consists of a humidifier and a second TSI 3563 nephelometer placed downstream of the first nephelometer. A one-hour programmable RH ramp (<40% to 85%) is applied to the air stream entering the second nephelometer. A two-parameter fit of the ratio of humidified to dried aerosol σ_{sp} is applied to each RH ramp deduce the RH dependence of σ_{sp} (Eq.3 of Titos et al., 2016). A similar fit is calculated for σ_{bsp} .”*
2. We now propagate estimated uncertainties in humidified σ_{sp} and σ_{bsp} to estimate uncertainties in RH-corrected SSA and g, for each season. We explain the methodology of these corrections in Sect. 3.1 via the following additions:
 - (a) *“Radiative transfer models typically only treat the scattering dependence when correcting ω_0 to ambient RH; and assume that absorption changes negligibly with RH. While this approach may or may not hold true for all aerosol types (ex: some organics, sulfur-coated soot), the dependence of σ_{ap} on RH is experimentally very difficult for all but laboratory studies (especially at high RH) conducted under very controlled conditions (Brem et al., 2012) and is ignored in our calculations. Thus, we only correct σ_{sp} to ambient RH in our corrections of ω_0 . Uncertainties in correcting σ_{sp} to ambient RH are due to uncertainties in (1) scattering coefficients measured by the dry and humidified aerosol nephelometers ($\Delta\sigma_{sp}=9.2\%$, Supplement to Sherman et al., 2015); and (2) RH inside the humidified nephelometer ($\Delta RH\sim 3\%$; Titos et al., 2016). Titos et al. (2016) used these values as inputs to a Monte Carlo simulation to estimate the uncertainty in the RH-corrected scattering coefficient as $\Delta\sigma_{sp}\sim 20\%$ (their Fig. 2b) for high-RH (>90%) and for moderately hygroscopic aerosols such as those observed at APP (Sherman et al., 2016b). We apply $\Delta\sigma_{sp}\sim 20\%$, along with uncertainty in dried aerosol absorption coefficient ($\Delta\sigma_{sp}=20\%$; Sherman et al., 2015), as inputs to Eq. S9 of supplement to Sherman et al. 2015 to calculate $\Delta\omega_0$. Single-scattering albedo uncertainty is larger for more absorbing aerosols and is zero for purely scattering aerosols ($\omega_0=1$). We use monthly median ω_0 values (Fig.5b) to calculate $\Delta\omega_0\sim 0.03$ for winter and surrounding months and $\Delta\omega_0\sim 0.02$ for summer and surrounding months (Table 2).”*
 - (b) *“Uncertainty in the calculated value of g at ambient RH arises due to uncertainties in the measured σ_{bsp} and σ_{sp} , each of which is subject to the same measurement*

uncertainties as outlined above. Sherman et al. (2015) reported a nearly identical uncertainty in dried aerosol hemispheric backscatter coefficient ($\Delta\sigma_{bsp}=8.9\%$) as for the scattering coefficient ($\Delta\sigma_{sp}=9.2\%$). This, along with the lack of published uncertainties in humidified $\Delta\sigma_{bsp}$ for similar experimental configurations as that deployed at APP, lead us to use the same uncertainty estimate for ambient-RH $\Delta\sigma_{bsp}$ as for ambient-RH $\Delta\sigma_{sp}$ ($\sim 20\%$). Inserting the ambient-RH uncertainties $\Delta\sigma_{bsp}$ and $\Delta\sigma_{sp}$ into Eq.S8 of supplement to Sherman et al. (2015) lead to hemispheric backscatter fraction uncertainty $\Delta b \sim 0.0085$, which in turn can be used along with the relation between g and b (Eq.3) to calculate $\Delta g = |\partial g / \partial b| \Delta b \sim 0.01$.”

3. We updated all measurement-based DRE uncertainty values in the manuscript, to reflect the updated measurement uncertainty values $\Delta\omega_0$ and Δg . The new uncertainty estimates do not give rise to any changes in the main results of the paper but we did need to make small modifications to the wording in places of the Results/Discussion and Summary/Conclusion sections (based on these changes)

Reviewer 1 Technical Comment 4: Authors present sensitivity of DRE to surface reflectance (S_R) at TOA and surface as $3.3Wm^{-2}$ and $2.7Wm^{-2}$ during June and $0:22Wm^{-2}$ and $0:20Wm^{-2}$ during December. What surprises me is the very small difference in S_R values at TOA and at surface. It is common knowledge that surface reflectivity will have very little effect on DRE at surface but can have significant effect at TOA. One can read reason for it in Chung (2012). In other words, a significant difference is expected between TOA and surface S_R values. See for example Figure 10 of Gadhavi and Jayaraman (2004) who have used similar approach and the same radiative transfer code (SBDART) to calculate DRE (they called it radiative forcing) as a function of AOD and surface reflectance. They have reported that when surface type changes from sea to sand (which is large change in surface reflectance) it causes a little change in DRE at surface but a large change in DRE at TOA for a fixed AOD. The values reported in the current manuscript may not be wrong but a thorough discussion needs to be included why their finding is at variance with others or the common knowledge. I believe such a discussion will add value to their manuscript as it will lead to better understanding of how non-aerosol parameter affects aerosol radiative forcing.

Authors' Response: We agree with the reviewer that more discussion of S_R and comparisons of our results with other papers such as Gadhavi and Jayaraman (2004) and McComiskey et al. (2008) could provide insight on the role of surface reflectance in aerosol DRE. To this end, we have made several related changes to the paper, enumerated below. We partially agree with the reviewer's assertion that “*It is common knowledge that surface reflectivity will have very little effect on DRE at surface but can have significant effect at TOA.*” Surface reflectivity can have either a large or small effect on DRE at TOA and at the surface. The difference between DRE (and DRE sensitivity) at the TOA and that at the surface is (for a fixed AOD) dependent on aerosol absorption and on the relative albedos of the atmosphere and underlying surface, with larger DRE differences (between TOA and surface) for more absorbing aerosols (low SSA) and brighter surfaces and smaller differences for less absorbing aerosols (higher SSA) and darker surfaces. Chung's Fig.5 depicts the case for very dark aerosols (SSA=0.19), which is mainly applicable to local sources of black carbon aerosols. Chung also states (first paragraph of their Sect.3) that “*The surface plays an important role in case of absorbing aerosols (i.e., aerosols*

with low SSA). As Fig. 5 shows, higher albedo (i.e., more reflection at the surface) increases aerosol absorption and thus aerosol forcing at the TOA as well as in the atmosphere. Higher albedo increases aerosol absorption because absorbing aerosols absorb not just the downward solar radiation but also the reflected upward radiation. Higher albedo also decreases aerosol scattering back to the space, further contributing to higher aerosol forcing at TOA. Ice, snow and desert have high surface albedo.)”. The aerosols at APP are ‘bright’ (SSA~0.91-0.96) and the surface is fairly dark (Fig(s).3 of manuscript) so the effects of changes in DRE due to small changes in surface reflectance (i.e. the DRE sensitivity S_R) will be relatively small, both at the TOA and at the surface. Our results are consistent with McComiskey et al. (2008), who also reported small differences (~10-20%) between S_R at the TOA and S_R at the surface for a continental US site, a tropical Pacific site, and a site in Alaska (their Fig.4).

Changes to paper:

1. The S_R values previously calculated (and referenced above by reviewer) were incorrectly calculated and we thank the reviewer for catching this! We now report sensitivity to surface albedo S_R as the slope of plot of DRE versus broadband (spectrally-averaged) R, not relative surface reflectance (as we had mistakenly calculated it). This facilitates more direct comparisons with McComiskey et al (2008) and Gadhavi and Jayaraman (2004), in addition to corrected DRE uncertainties due to the use of the proper S_R values. There are no major changes in the main results of the paper but small changes in the DRE uncertainty values and larger absolute S_R values, that are in much better agreement with McComiskey et al. (2008).
2. We modified the following passages to Sect. 4.3, more clearly explaining how S_R is calculated.
 - (a) “For the sensitivity S_R , we scale the entire spectral surface reflectance curve (Figs.3) by proportionally scaling the input surface type coefficients supplied to SBDART (Fig.3), to vary the broadband (250-4000nm) surface reflectance R (Figs. 6d and 7d). For example, doubling both the sand and vegetation coefficient values supplied to SBDART scales the entire surface reflectance curve by the same amount, thereby doubling the base-case value of broadband R in Table 1.”
 - (b) “The base case R values in Table 1 are the broadband surface reflectance corresponding to the monthly mean spectral surface reflectance curves (Figs.3). We then vary the independent variables ρ_i individually about these base case values (Table 1) to generate the ‘seasonal’ DRE versus ρ_i curves. We evaluate $S_i = \partial (\text{DRE}) / \partial \rho_i$, at base case ρ_i value, as the regression slope of the five points on each side of the base case value
3. We re-made the plots of S_R (Figs. 6c and 7c) so that they now plot curves of DRE versus spectrally-averaged R (Change 1 above), not DRE versus relative surface reflectance.
4. We compare our S_R values to those of McComiskey et al (2008) and Gadhavi and Jayaraman (2004) and discuss the results in Sect. 5.2, with the paragraph shown below.

We also added curves of DRE versus AOD for the same surface types as , Gadhavi and Jayaraman (2004), to the supplement of our paper. This allowed for a more direct comparison of our S_R results with theirs.

“Relatively low sensitivity of DRE to surface reflectance at APP during the study period is seen by the low S_R values, which range from 17 Wm^{-2} (14 Wm^{-2}) at the TOA (surface) during JUN to 2.0 (1.8) Wm^{-2} at TOA (surface) during DEC, per unit change in R (Table 3). Surface reflectance changes very little during vegetative summer months and large changes within a given season typically only occur for extended snow cover during some winter months, when S_R is very small. Our S_R values for SEP (9.1 Wm^{-2} at TOA and 7.7 Wm^{-2} at surface, per unit change in R) are very close to those reported by McComiskey, et al. (2008) for the SGP site (their Fig.4). While TOA S_R values in our study and that of McComiskey et al. (2008) are only ~10-20% larger than S_R values at the surface, Gadhavi and Jayaraman (2004) reported much higher DRE sensitivity to surface type at the TOA than at the surface in Antarctica. As part of their analysis, Gadhavi and Jayaraman (2004) plotted both TOA and surface DRE versus AOD for different surface types (snow, seawater, sand, and vegetation), using the same SBDART radiative transfer code used in our study and that of McCommiskey et al. (2008). Gadhavi and Jayaraman (2004) reported similar TOA DRE sensitivities to changes in surface albedo as we do. Their TOA DRE changed by $\sim 10 \text{ Wm}^{-2}$ as they changed the surface type from all sea water to all snow (their Fig.10, for $\text{AOD}=0.1$), which represents close to a unit change in surface albedo. However, their corresponding change in surface DRE was only $\sim 3 \text{ Wm}^{-2}$. To determine whether the much smaller surface S_R reported by Gadhavi and Jayaraman (2004) is due to the method used to report them, we plot similar curves of DRE versus AOD for each of the four surface types (Fig.S3 of supplement to this paper). The differences of DRE calculated for pure snow and pure seawater (at $\text{AOD}=0.10$) are similar to our S_R values derived using the sensitivity curves (Fig.5), both at the TOA and at the surface. This leads us to speculate that the smaller surface S_R reported by Gadhavi and Jayaraman (2004) could be due to differences in aerosol properties and/or their spectral dependence supplied to the SBDART model, or else on how spectral surface reflectance was parameterized. Gadhavi and Jayaraman (2004) did not state the spectral ω_o used as input to the RTM and aerosol absorption can have a very large effect on DRE sensitivity to the underlying surface type (Chung, 2012).”

Reviewer 1 Technical Comment 5: Page 35 caption of Fig. 1: Longitude number of the site should have suffix “W”.

Authors’ Response: Thanks for catching that!

Change to Paper: We have corrected the longitude in Fig.1, changing “N” to “W”,